

POTENTIAL OF KAOLIN- PALM OIL FUEL ASH MIXTURE AS  
SUSTAINABLE LANDFILL LINER MATERIAL

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A thesis submitted in fulfilment of the  
requirements for the award of the degree of  
Doctor of Philosophy (Civil Engineering)

Faculty of Civil Engineering  
Universiti Teknologi Malaysia

JANUARY 2014

**In the Name of ALLAH, the Most Gracious, the Most Merciful.**

To:

My Father: Late Prince (Haj.) Mustapha Oyeleke (1917-1983)

My Mother: Late Haja Sabtiyu Oyeleke (1920-2004), who gave me an excellent upbringing and cherished me with love and tenderness. May Almighty Allah subhanahu wa ta 'la continue to grant their souls with perfect peace and eternal rest, and may He also count them and us among His obedient servants on the day of judgment, (amin).

My Heartthrob: Haja Khadijat Kuburah Bin Mohd.Jimoh, for her understanding and unalloyed cooperation.

My children:

Prince:

Mubarak Adeyemi (1994)

Khalid Adeyinka (1996)

Saleem Adegoke (1999)

Ateeq Adeboye (2001), and

Princess:

Kabirah Abike (2005), for their patience and perseverance when I left them to pursue this noble course, at the stage when my attention is highly in need.

## ACKNOWLEDGMENT

Firstly, I would like to express my profound gratitude to Almighty Allah (S.W.T) for showing me His guidance and blessing, as He has enable me complete this Ph.D thesis. May the peace and blessings of Almighty Allah (S.W.T) be upon our Noble Prophet Muhammad (S.A.W) for he is the unlettered messenger who has brought light to mankind and take them out of total darkness and ignorance. My sincere thanks also go to several institutions and persons numerous to be mentioned here.

Secondly, my deepest gratitude is directed to my able and astute supervisors, Dr. Mohd. Badruddin Bin Mohd. Yusof for his expertise advice, dedication, guidance, constructive criticisms, textual language checking system, and in thesis outlining. He is always available for me during the course of this research. I also want to sincerely thank my co-supervisors, in persons of Prof. Dr. Mohd. Razman Bin Mohd. Salim and Associate Prof. Kamarudin Ahmad for their invaluable suggestions, spirit of motivation and extensive knowledge which have contributed immensely to the success of this thesis. They have assisted me greatly in the collections of pertinent literatures, effective interpretation of all textual materials used and understanding of the system of note-taking, and finally, synthesizing from different sources. This has greatly contributed to the originality of this thesis.

In the absence of sustained financial support and in particular emotional stability, it would have been difficult for me to complete my thesis. For these two viable and important Ph.D parameters, I would like to clearly offer my deepest gratitude to the Universiti Teknologi Malaysia (UTM), for the IDF tuition scholarship offered for two semesters as financial leverage during the course of my

program. My thanks also go to the Librarians and other supporting staffs of Perpustakaan Sultanah Zahariah (PSZ), UTM Library for their assistance in making the relevant literatures and serene library environment available for this research.

Finally, I will like to extend my sincere appreciation to the following distinguished personalities and organizations for their unalloyed cooperation and unwavering supports, throughout the period of my studies at UTM. They are Haj. Abdel Yekeen Oyeleke; Haj. Mikhaeel Tayo Oyeleke; Haj. Semee Oyeleke; Haj. Taofeeq Oyeleke; Prince Abd. Hameed Adeyemi Onifade; and Muthashiru Kolawole. Others are Adewale Adeyemo; Sarafadeen Kolawole; Arc. Kalejaiye Abdurazak; Femi Olateru; Prince Munir Oyewale Sholadoye; Abdulkareem Ayinla; Taliat Yusuf; and the Oni Ira of Ira Land, His Royal Highness Abdulwahab Oyetero; the entire sons and daughters of Ira Land; the Management of Kaduna Polytechnic, Built Environment Ltd, In-depth Engineering and Osbark Engineers Ltd. May Allah reward all of them for their opened or confined contributions and sincere support towards achieving this noble objective (amin).

## ABSTRACT

Contamination of groundwater due to leachate percolating through landfill liner is a potential hazard to human health and the environment. Effective mitigation approaches and imperative research on alternative liner materials could reduce contamination. In this study, an investigation on the potential of kaolin premixed with pulverized palm oil fuel ash (PPOFA) as an alternative mineral liner material is proposed. The investigation was carried out in two stages. Initially, physico-chemical and mechanical characterizations were performed on these materials separately. Then, PPOFA was mixed in stages of 0, 10, 20, and 30 percent by dry mass of the kaolin and were compacted using Standard Proctor. Result from the compaction test showed that 15 percent of PPOFA was found to be the optimum dose for the formulated liner matrix. In the second stage, material characterizations were repeated on the formulated liner matrix followed by performance criteria tests. These tests included heavy metal removal efficiency, chemical sorption, leaching, hydraulic conductivity and column tests. The criteria examined were compared with the standards and previous related research. All the tests were conducted without pH adjustment and were duplicated. Based on sorption tests, Freundlich and Langmuir isotherm models were developed and validated using linear regression and correlation coefficient  $R$ . The performance tests recorded high metal ion removal efficiencies of 59 to 99 percent at solution equilibrium pH ranging from 7.64 to 3.00. Freundlich sorption capacities ranged from 0.2063 to 25.64, while the Langmuir monolayer sorption capacity,  $\beta$  ranged from 22.3714 to 52.6316  $\text{mgg}^{-1}$ . Besides that, the modelled Freundlich ( $q_{eFred}$ ) and Langmuir ( $q_{eLang}$ ) isotherm were relatively well fitted to the experimental ( $q_{expt}$ ), with correlation coefficients ( $R$ ) greater than 0.84. Leaching occurred at very low concentrations ranging from 0.003 to 0.19  $\text{mgL}^{-1}$  and hydraulic conductivity tests obtained values ranging from 3.14 to  $3.66 \times 10^{-6} \text{ cms}^{-1}$ . Finally, the column test showed that the hydrodynamic dispersion coefficients ( $D_L^*$ ) and retardation factors ( $R_d$ ) ranged between  $1.16 \times 10^{-3}$  to  $1.11 \times 10^{-5} \text{ cm}^2\text{s}^{-1}$  and 17 to 337 respectively. The proposed kaolin-PPOFA liner matrix has been proven to be a promising alternative liner material in ameliorating groundwater contamination from the leachate generated in the landfill.

## ABSTRAK

Pencemaran air bumi yang disebabkan oleh penelusan air larut lesap melalui pelapik tapak pelupusan adalah berpotensi membahayakan manusia dan alam sekitar. Pendekatan pencegahan yang berkesan dan penyelidikan yang imperatif mengenai bahan-bahan pelapik ini dapat mengurangkan pencemaran. Menerusi kajian ini, satu penyiasatan telah dicadangkan ke atas potensi campuran kaolin dengan serbuk abu bahan bakar kelapa sawit (*PPOFA*) sebagai alternatif kepada bahan mineral pelapik. Penyelidikan ini telah dijalankan dalam dua peringkat. Pada mulanya, pencirian kimia-fizikal dan mekanikal telah dilakukan ke atas bahan-bahan ini secara berasingan. Kemudian, bahan *PPOFA* telah dicampurkan pada tahap 0, 10, 20, 30 peratus jisim kering kaolin dan dipadatkan menggunakan kaedah Piawaian *Proctor*. Melalui ujian pemadatan, *PPOFA* 15 peratus merupakan dos optimum untuk matriks pelapik yang telah diformulasikan. Pada peringkat kedua, pencirian bahan telah diulangi pada matriks pelapik yang diformulasikan diikuti dengan ujian prestasi kriteria. Ujian-ujian ini termasuklah kecekapan penyingkiran logam berat, penyerapan kimia, larut lesap, kekonduksian hidraul dan ujian turus. Kriteria yang dianalisa telah dibandingkan dengan piawaian dan penyelidikan sebelumnya yang berkaitan. Kesemua ujian telah diulang sebanyak dua kali tanpa mengubah nilai pH. Berdasarkan ujian penyerapan, model lengkung sesuhu Freundlich dan Langmuir telah dihasilkan dan disahkan menggunakan kaedah regresi lurus dan pekali korelasi  $R$ . Prestasi ujian merekodkan nilai kecekapan penyingkiran ion logam yang tinggi daripada 59 hingga 99 peratus pada keseimbangan pH larutan antara 7.64 hingga 3.00. Kapasiti erapan Freundlich bernilai antara 0.2063 hingga 25.64, manakala nilai kapasiti erapan ekalapisan Langmuir,  $\beta$  adalah antara 22.3714 hingga 52.6316  $\text{mgg}^{-1}$ . Di samping itu, model lengkung sesuhu Freundlich ( $q_{e\text{Fred}}$ ) dan Langmuir ( $q_{e\text{Lang}}$ ) yang dihasilkan memberi nilai yang menghampiri nilai eksperimen ( $q_{\text{expt}}$ ), dengan pekali korelasi ( $R$ ) lebih besar daripada 0.84. Larutlesapan berlaku pada kepekatan yang sangat rendah iaitu antara 0.003 hingga 0.19  $\text{mgL}^{-1}$  dan ujian kekonduksian hidraul memberi nilai antara 3.14 hingga  $3.66 \times 10^{-6} \text{ cms}^{-1}$ . Akhir sekali, ujian turus menunjukkan nilai pekali serakan hidrodinamik ( $D L^*$ ) dan faktor perencatan ( $R_d$ ) yang masing-masing bernilai antara  $1.16 \times 10^{-3}$  ke  $1.11 \times 10^{-5} \text{ cm}^2\text{s}^{-1}$  dan 17 ke 337. Matriks pelapik kaolin-PPOFA yang dicadangkan telah dibuktikan sebagai alternatif bahan pelapik yang berpotensi dalam membendung pencemaran air bumi dari proses larut lesap yang dihasilkan di tapak pelupusan.

## TABLE OF CONTENTS

<b>CHAPTER</b>	<b>TITLE</b>	<b>PAGE</b>
	<b>DECLARATION</b>	ii
	<b>DEDICATION</b>	iii
	<b>ACKNOWLEDGMENT</b>	iv
	<b>ABSTRACT</b>	vi
	<b>ABSTRAK</b>	vii
	<b>TABLE OF CONTENTS</b>	viii
	<b>LIST OF TABLES</b>	xxi
	<b>LIST OF FIGURES</b>	xxiv
	<b>LIST OF ABBREVIATIONS</b>	xxix
	<b>LIST OF SYMBOLS</b>	xxxix
	<b>LIST OF APPENDICES</b>	xxxiv
<b>1</b>	<b>INTRODUCTION</b>	<b>1</b>
1.1	Background of the Problem	1
1.2	Statement of the Problem	5
1.3	Aim and Objectives of the Research	8
1.4	Scope of the Research	8
<b>2</b>	<b>LITRATURE REVIEW</b>	<b>11</b>
2.1	Introduction	11
2.2	The Formation of Primary and Secondary Clay Minerals	11



2.2.1 The Basic Building Blocks of Kaolinite Mineral	12
2.2.1.1 The Formation of Unit Alumina and Silica Sheet	13
2.2.1.2 Net Positive Charge per Aluminium Octahedral Unit	14
2.2.1.3 Net Negative Charge per Silicon Tetrahedral Unit	14
2.2.1.4 The Formation of Kaolinite Structural Unit from the Unit Alumina and Sheets	14
2.2.1.5 The Formation of Kaolin Crystal from Kaolinite Structural Units	15
2.2.2 Physical Properties of Kaolin	16
2.2.3 Mechanical Properties of Kaolin	17
2.2.4 Chemical and Geotechnical Properties of Kaolin	17
2.2.4.1 Isomorphous Substitutions Processes and Formation of Binding Sites on Kaolinite	18
2.2.4.2 The Process of Cation-exchange in Kaolinitic Clay	20
2.2.4.3 Clay-water-ion Interaction System	20
2.2.4.4 Ionic Distribution in Clay-leachate Environment	21
2.2.4.5 Diffuse Double Layer in Hydrated Clay Mineral	21
2.2.5 Sourcing and Availability of Kaolin in Malaysia	22
2.2.5.1 General usage and Application	23
2.2.6 The Production and Challenges of Palm Oil Mill Biomass in Generating Sustainable Green Energy in Malaysia	24
2.2.6.1 The Production of POFA from Palm Oil Mill Biomass Residue	25
2.2.6.2 Statistics of POFA Generation in Malaysia Based on Quantity of Palm Oil Produced	

	in 2012 and 2013	25
	2.2.6.3 Chemical Composition of POFA	29
	2.2.6.4 Cementitious and Adsorbent Reactivity of PPOFA	30
	2.2.6.5 Application of POFA in Environmental Engineering	31
2.3	Review of Imperial Literatures on Liner Minerals	32
	2.3.1 Current Materials used as Liner in Modern Landfills	32
	2.3.1.1 Soil Liner	32
	2.3.1.2 Geomembrane Liner	34
	2.3.1.3 Geosynthetic Clay Liner	34
	2.3.2 Alternative Liner Materials	36
	2.3.2.1 Bottom and Fly ash from Coal Industry	36
	2.3.2.2 Rubber and Bentonite added Fly ash	37
	2.3.2.3 Bottom Ash from Sludge Cake as Liner Material	37
	2.3.2.4 Composting of Municipal Solid Waste as Liner Material	38
	2.3.3 Mechanical Activation of Geo-mineral liner Material	38
	2.3.4 Hydraulic Conductivity of Mineral Landfill Liner	39
	2.3.4.1 Compaction and Hydraulic Conductivity of Earthen Liner	40
	2.3.4.2 Effect of Biomass Ash on the Hydraulic Conductivity of Mineral Liner	41
	2.3.4.3 The Role of DDL on Hydraulic Conductivity	42
	2.3.5 Sorption and Retention Capacity in Mineral Liner	43
	2.3.6 Total Mass Flux of Contaminant Species	47
	2.3.6.1 Application of Contaminant Transport Model in Mineral Liner	48

2.3.6.2	Analytical Solution to the Governing Equation of Contaminant Transport in Mineral Liner	49
2.3.6.3	Longitudinal Hydrodynamic Dispersion and Retardation Coefficients of Dissolved Solutes in Landfill Leachates	51
2.3.7	Kaolinitic Clay as Adsorbent and Landfill liner	52
2.3.7.1	Calcination of Kaolinitic Clay and Pozzolanicity	54
2.3.8	POFA as Pozzolanic and Adsorbent Material	54
2.3.8.1	POFA as Pozzolanic Material in Landfill Liner	55
2.3.8.2	POFA as Adsorbent in Removal of Dye	55
2.3.8.3	POFA as Adsorbent in Removal of Toxic Metal Ion Solution	56
2.3.9	Leachability of Industrial Waste Materials	57
2.4	Overview of Landfill Technology	58
2.4.1	Planning of Sanitary Landfill	58
2.4.2	Preparation of Landfill Site and Installation of Monitoring Devices	58
2.4.2.1	Site Excavation	59
2.4.2.2	Installation of Monitoring Devices and Landfill Liner	59
2.4.3	Operation of Landfill	59
2.4.3.1	The Placement of Solid Wastes	60
2.4.3.2	Leachate Generation	60
2.4.3.3	Leachate collection system	61
2.4.3.4	Landfill Gas Generation and Installation of Gas Recovery Trench	61
2.4.3.5	Landfill Liners	61
2.4.4	Landfill Closure and Post-closure Control	62
2.4.4.1	Final Cover Material	62
2.4.4.2	Maintenance of Settled Final Surface	62

2.4.5	Landfill Leachate Management	63
2.4.5.1	Leachate Recycling	63
2.4.5.2	Leachate Evaporation	65
2.4.5.3	Leachate Treatment	65
2.4.5.4	Discharge to Municipal Wastewater Collection Systems	66
2.4.5.5	Integrated Leachate Management System	67
2.4.6	Pollution Problems at Landfill Site	67
2.4.6.1	Atmospheric	68
2.4.6.2	Hydrological	68
2.4.6.3	Other Pollution Problems at Landfill Site	69
<b>3</b>	<b>MATERIALS AND METHODS</b>	<b>70</b>
3.1	Introduction	70
3.2	Production of Crude Palm Oil in Tai Tak Palm Oil Mill	70
3.3	Generation of Raw POFA as Solid Waste Material in Tai Tak Palm Oil Mill	73
3.4	Sourcing, Sampling and Pre-treatment of Raw POFA	74
3.4.1	Collection and Sampling of the Raw POFA	75
3.4.2	Laboratory Pre-treatment of the Raw POFA	75
3.4.3	Sieving of the Raw POFA after Oven Dried	76
3.4.4	Mechanical Activation of the Sieved POFA	79
3.4.4.1	Hammer Milling Process	80
3.4.4.2	Ball Milling Process	81
3.5	Sourcing and Pre-treatment of the Industrial Kaolin	82
3.5.1	Sourcing of the Industrial Kaolin	82
3.5.2	Pre-treatment Process of the Industrial Kaolin	83
3.6	Physical Characterization of the Test Materials	83
3.6.1	Particle Size Gradation	83
3.6.2	Particle Density Analysis	85
3.7	Chemical Characterization of the Test Materials	86
3.7.1	Determination of Mass Loss on Ignition and Unburnt Carbon Content	87

3.7.2	Determination of Scanned Particle Morphology	88
3.7.3	Determination of the BET Surface Area and Pore Characteristics	89
3.7.4	Chemical and Elemental Characterization	90
3.7.5	Determination of Mineral Compositions	90
3.7.6	Determination of pH and EC	92
3.7.7	Determination of Exchangeable Cations and Cation Exchange Capacity	93
3.8	Mechanical Characterization of the Test Materials	95
3.8.1	Determination of Atterberg Consistency Values	95
3.8.1.1	Determination of Liquid Limit by Cone Penetrometer (dry-to- wet method)	96
3.8.1.2	Determination of Plastic Limit	96
3.8.1.3	Calculation of the Plasticity Index	97
3.8.2	Classification of the Materials Based on the BSCS Plasticity Chart	98
3.8.3	Compaction Characteristics of the Industrial Kaolin Modified with the PPOFA at varying Percentage Dosage Level	98
3.8.3.1	Determination of Initial Moisture Content	99
3.8.3.2	Standard Proctor Compaction Test	99
3.8.3.3	Plotting of the Moisture-density Relationship Curve and obtaining the Corresponding Value of Maximum Dry Density and Optimum Moisture Content	101
3.8.3.4	Graphical Estimation of the PPOFA Dosage Level Based on Compaction Tests	101
3.9	Batch Adsorption Equilibrium Test for the Formulated Geo-liner Material	102
3.9.1	Preparation of the Aqueous Metal Solutions	102
3.9.2	Sequential Batch-test for Fixing the	

	Liner-to-leachate Ratio	104
3.9.3	Material Set-up for the Batch Adsorption Isotherm Test	105
3.9.4	Experimental Process for the Batch Adsorption Isotherm Test	106
3.9.5	Determination of Equilibrium Concentration and pH of the Test Specimens	107
3.9.6	Heavy Metal Removal Efficiency and Sorption Capacity of the Formulated Geo-mineral Liner Material	109
3.9.7	Adsorption Parameters from Freundlich and Langmuir Model	110
3.9.8	The Regression Line and Parameters for Calculating the Freundlich Sorption Isotherm Model Parameters	110
3.9.9	The Regression Line and Parameters for Calculating the Langmuir Sorption Isotherm Model Parameters	111
3.10	Leaching Test of the PPOFA, Industrial kaolin and the Formulated Geo-mineral Liner Material	112
	3.10.1 Batch Leaching Performance Test	112
3.11	Hydraulic Conductivity Test for the Formulated Geoliner Material	114
	3.11.1 Fabrication of Acrylics Test Cells	114
	3.11.2 Preparation of Test Materials	115
	3.11.3 Proportioning and Storage of the Prepared Liner Material	116
	3.11.4 Extrusion of air from Voids in Liner Material	117
	3.11.5 Material Compaction Process	118
	3.11.6 Initiating Pozzolanic Reaction in the Liner Columns	119
	3.11.7 Permeameter Assemblage	120
	3.11.8 Establishing Hydraulic Conductivity of the	

	Compacted Liner Material Permeated with De-aired DW water (the falling head permeability method)	122
3.12	Column Diffusion-retardation Test	123
	3.12.1 Preparation of the Four Single Metal Species in Aqueous Solutions	123
	3.12.2 Column Assemblage and Identification System	124
	3.12.3 Column Diffusion-retardation Test	125
	3.12.4 Seepage Velocity of the Column Liner assigned to each the Heavy Metal Solution	126
	3.12.5 Analytical Solution to Determine the Longitudinal Hydrodynamic Dispersion Coefficient and Retardation Factor	128
	3.12.5.1 Proposed Analytical Solution to the Contaminant Transport Model	128
	3.12.5.2 The use of Modified Analytical Solution to Determine the Contaminant Transport Parameters	128
	3.12.5.3 Optimization of the Proposed Model to Determine the Longitudinal Hydrodynamic Dispersion Coefficient and Retardation Factor	130
	3.12.5.4 The use of the Excel's Solver as Optimization Tool	130
3.13	Performance Evaluation Criteria	131
	3.13.1 Evaluation Based on the Heavy Metal Removal Efficiency and Sorption Capacity	131
	3.13.2 Evaluation Based on the Leachability of the Heavy Metals Tested	131
	3.13.3 Evaluation Based on the Hydraulic Conductivity	132
	3.13.4 Evaluation Base on the Longitudinal Hydrodynamic Dispersion Coefficient and Retardation Factor	132

3.14	Calibrations of the Laboratory measuring Instrument and Equipment for Reliability of Data	133
<b>4</b>	<b>RESULTS AND DISCUSSIONS</b>	<b>134</b>
4.1	Introduction	134
4.2	Physical Characterization of the PPOFA and the Industrial Kaolin	135
	4.2.1 Visual and Manual Classifications	135
	4.2.2 Particle Size Gradation Parameters	138
	4.2.3 Particle Density Analysis	141
4.3	Chemical Characterization of the PPOFA and the Industrial Kaolin	142
	4.3.1 Mass Loss on Ignition and Unburn Carbon Content	142
	4.3.2 Scanned Particle Morphological Analysis	144
	4.3.2.1 Surface Scanned Particle Morphological Structure	144
	4.3.3 Surface Area and Pore Characteristics	146
	4.3.3.1 Surface Area Characteristics	147
	4.3.4 Chemical and Elemental Compositions	148
	4.3.5 Mineral Compositions	151
	4.3.6 pH and EC Determination	155
	4.3.7 Exchangeable Cations and Cation Exchange Capacity	156
4.4	Mechanical Characterization of the PPOFA and the Industrial kaolin	158
	4.4.1 The Atterberg Consistency Limit States and Plastic Index	158
	4.4.2 Classification Based on BSCS Plasticity Chart	159
	4.4.3 Compaction Characteristics of the Industrial Kaolin Modified with the PPOFA at Varying Percentage Dosage	159
	4.4.3.1 Moisture-density Relationship Curves and	



	Corresponding values of Maximum Dry Density and Optimum Water content at Varying Percentage PPOFA Dosage	159
	4.4.3.2 The Effect of Percentage PPOFA Dose on Maximum Dry Density	161
	4.4.3.3 The Effect of Percentage PPOFA Dose on Optimum Moisture Content	162
	4.4.3.4 The Effect of Kaolin – PPOFA Mixture on the Position of the 100 % Saturation Line	163
	4.4.3.5 Graphical Estimation of the Percentage PPOFA Dose for the Design of the Landfill Liner Material	163
	4.4.3.6 Implicit Deduction for the Actual PPOFA Dosage Level in the Liner Material	165
4.5	Physical Characterization of the Formulated Geo-mineral liner Material	166
	4.5.1 Visual and Manual Classifications	167
	4.5.2 Particle Size Gradation Parameters	168
	4.5.3 Particle Density Analysis	170
4.6	Chemical Characterization of the Formulated Geo-liner Material	171
	4.6.1 Mass Loss on Ignition and Unburn Carbon	171
	4.6.2 Surface Scanned Particle Morphological Structur	171
	4.6.3 Surface Area Characteristics	173
	4.6.4 Characterization of the Pore Parameters	173
	4.6.5 Chemical and Elemental Compositions	174
	4.6.6 Mineral Compositions	175
	4.6.7 pH and EC Determination	176
	4.6.8 Exchangeable Cations and Cation Exchange Capacity	178
4.7	Mechanical Characterization of the Formulated Geo-liner Material	178

4.7.1	The Atterberg Consistency Limit States and Plastic Index	179
4.7.2	Classification Based on BSCS Plasticity Chart	180
4.8	Performance Evaluation Criteria for the Formulated Geo-liner Material	181
4.8.1	Heavy Metal ion Removal Efficiency	181
4.8.2	Initial Parameters of the Metal ion Solutions	181
4.8.2.1	The Effect of Initial Heavy Metal Concentration on the Equilibrium $pH_e$ and $EC_e$	182
4.8.2.2	Trend in the Metal Removal Efficiency	183
4.8.2.3	Factors Attributed to the Relatively low Metal Removal Efficiency at Relatively High Metal Concentration	185
4.8.2.4	Enhancing the Overall Liner Performance Efficiency	187
4.9	Adsorption Capacity	189
4.9.1	Feasible Sorption Isotherm Data Points Adopted for plotting the Regression Lines	189
4.9.2	The Regression Line and Parameters for Calculating the Freundlich and Langmuir Sorption Isotherm Model Parameters	190
4.9.3	The Generated Linear Regression Model used to calculate the Freundlich and Langmuir Sorption Isotherm Model Parameters and the Coefficient of Determination	192
4.9.4	Freundlich and Langmuir Sorption Isotherm Parameters and Models for the Geo-liner Material with respect to the Heavy Metal Species	193
4.9.5	The Effect of Protonation Reaction of $H^+$ on the Sorption of the $Cd^{2+}$	195
4.9.6	Comparison of the Experimental and Modeled Mass of the Heavy Metal ions Sorbed per unit	

	Dry Mass of the Liner Material	196
4.9.7	Validating Sorption Responses from the Established Freundlich and Langmuir Isotherm Model by Curve Fitting and Correlation Coefficient	197
4.9.7.1	Validating the Freundlich and Langmuir Isotherm Model from the Output $q_{eFred}$ and $q_{eLang}$ in relation to the Experimental $q_{expt}$ by the Degree of Curve Fitness	198
4.9.7.2	Correlation Co-efficient of the Model Output $q_{eFred}$ and $q_{eLang}$ in relation to the Experimental $q_{expt}$	201
4.10	Leaching Test	202
4.10.1	Batch Leaching Procedure Analysis	202
4.11	Hydraulic Performance of the Compacted Geo-liner Material	203
4.11.1	Geotechnical Parameters	204
4.11.2	Established Steady Flow Condition	206
4.11.3	Hydraulic Conductivity	207
4.11.4	Statistical Inferences Drawn from Hydraulic Conductivity Test	209
4.11.5	Factor Responsible for the Recorded Relatively Lower Dry Density	209
4.11.6	Factors Attributed to the Relatively Low Hydraulic Performance	210
4.12	Heavy Metal Diffusion and Retardation Performance Analysis	213
4.12.1	Chemical Properties of the Prepared Heavy Metal Solutions	214
4.12.2	Seepage Velocity of the Column Liner Assigned to each of the Heavy Metal Solution	215
4.12.3	Chemical Characteristics of the Effluent with variation in the Cumulative Pore Volume Flow	216

4.12.4	The Longitudinal Hydrodynamic Dispersion Coefficient and Retardation Factor	218
4.12.4. 1	The Longitudinal Hydrodynamic Dispersion Coefficient of the Metal Solute in the Liner Material	219
4.12.4.2	The Retardation Factor of the Metal Solute in the Liner Material	220
<b>5</b>	<b>CONCLUSIONS AND RECOMMENDATIONS</b>	<b>222</b>
	5.1 Introduction	222
	5.2 Recommendations	225
	<b>REFERENCES</b>	<b>227</b>
	Appendices A-T	243-280

## LIST OF TABLES

TABLE NO.	TITLE	PAGE
2.1	Monthly Production of Oil Palm Products for the Month of August 2012 and 2013 (Tonnes)	27
2.2	Annual and Forecast of crude Oil Production for the 2012 and 2013 (Tonnes)	28
2.3	Chemical composition of OPC in comparison with pulverized and thermally treated pulverized POFA	30
2.4	Characteristics of raw leachate from Seelong landfill in Johor-Malaysia	64
3.1	Test sieve designations and sizes to BS standard used for the POFA sieving	78
3.2	Targeted and corresponding actual aqueous solutions prepared for Zn <sup>2+</sup> ion solutes (Temp. = 26.50 ± 0.01 °C)	104
3.3	Geotechnical parameters of the compacted liner columns	119
3.4	Chemical characteristics of the metal solutes	124
3.5	Hydraulic parameters of the formulated liner material	127
4.1	Visual and manual classifications of the test materials	137
4.2	Salient particle gradation parameters for the test materials	140
4.3	BET fineness and pore characteristics of the tested materials	147
4.4	Chemical and elemental composition of the PPOFA	149
4.5	Chemical and elemental composition of the industrial	

	kaolin	151
4.6	Mineral compositions of the PPOFA and the industrial kaolin	153
4.7	Exchangeable cations and cation exchange capacity of the tested materials	157
4.8	Optimum water content and corresponding maximum dry unit weight recorded at varying percentage PPOFA dose in the trial kaolin-PPOFA mix	161
4.9	Visual and manual classifications of the formulated kaolin-PPOFA liner material	168
4.10	Salient particle gradation parameters for the formulated kaolin-PPOFA liner material	170
4.11	Chemical characteristics of the formulated kaolin-PPOFA liner material	172
4.12	Chemical and elemental composition of the formulated kaolin-PPOFA liner material	174
4.13	Mineral compositions of the formulated kaolin-PPOFA liner material	176
4.14	pH, Ec and Exchangeable cation concentration of the formulated kaolin-PPOFA liner material	177
4.15	Linearized regression models used to determine the parameters of the Freundlich isotherms for the sorbed metal ions	191
4.16	Linearized regression models used to determine the parameters of the Langmuir isotherms for the sorbed metal ions	191
4.17	Linear regression model used in developing the Freundlich and Langmuir isotherm model parameters and the $R^2$ for single-metal solution	193
4.18	Comparison of Langmuir and Freundlich sorption isotherm parameters and model for the single-element system	194
4.19	Experimental and modeled mass of heavy metal ions	

	sorbed per unit dry mass of the formulated mineral liner material	197
4.20	Correlation coefficient of $q_{eFred}$ and $q_{eLang}$ in relation to $q_{expt}$ at 5% significant level	201
4.21	BTP results of PPOFA, kaolin, and kaolin-PPOFA mixtures	203
4.22	Geotechnical parameters of the compacted liner columns	205
4.23	Established steady-state flow condition for DI water through the liner columns	207
4.24	Hydraulic parameters of the formulated kaolin-PPOFA liner material	215
4.25	The longitudinal hydrodynamic dispersion coefficients and retardation factors obtained from column tests for the single-metal solutions	219

## LIST OF FIGURES

<b>FIGURE NO.</b>	<b>TITLE</b>	<b>PAGE</b>
2.1	The basic structural units of kaolinite minerals: a) aluminum ion enclosed by octahedron of hydroxyls; b) silicon ion surrounded by tetrahedron of oxygen atoms	12
2.2	Structural network of octahedral forming an alumina sheet	13
2.3	Hexagonal network of tetrahedral forming a silica sheet	13
2.4	The 1:1 kaolinite structure layer a) molecular structure b) Schematic presentation	15
2.5	Isomorphism in kaolinite clay mineral leads to net negative charge on clay particles	19
2.6	Distribution of ions adjacent to charged clay surface according to the concept of diffuse double layer	22
3.1	Flow chart outline of laboratory activities mile stones for the research	71
3.2	The processes of extraction of crude palm oil from fresh fruit bunch, in Tai Tak oil milling industry	72
3.3	Mixture of bottom-ash and fly-ash deposited at the repository of Tai Tak Palm Oil Mill firm	74
3.4	Collection of the raw POFA after sampling from the stock pile waste	75
3.5	Raw POFA under process of open sun-dry	76



3.6	Set of test sieve used for effective capture of the raw POFA in progress	77
3.7	The sieved POFA content retained on 75 $\mu\text{m}$ and 38 $\mu\text{m}$ and in the receiving pan ready for grinding	78
3.8	POFA being removed from the Los Angeles abrasion machine after a milling process completed	80
3.9	FRITSCH planetary mill in operation	81
3.10	Bags of kaolin stacked at Tai Tak oil mill firm	82
3.11	PSA-CILAS 1180 particle size analyser	84
3.12	Vacuum pump for de-airing of specimens during determination of particle density	85
3.13	Specimens in the ELE electric muffle furnace for the LOI analysis	87
3.14	Energy Dispersive X-Ray Analyser for the structural scanned electron morphology test	89
3.15	Bench-top EDXRF-Minipal 4 X-ray spectrometer-PW4030 used in determination of oxides in the test materials	91
3.16	Brucker D8 Advanced X-Ray diffractometer used for the mineral detection test	91
3.17	PPOFA and kaolin-PPOFA geo-liner mixture prepared for pH and EC determination	92
3.18	Triplicate specimens of the industrial kaolin prepared for pH and EC determination	92
3.19	Kaolin-PPOFA prepared for the single run of independent standard Proctor compaction test	100
3.20	A set of replicated kaolin-PPOFA geo-liner specimen prepared for the batch isotherm test	106
3.21	HOTECH variable speed orbital shaker in progress single-batch extraction process	107
3.22	Duplicated kaolin-PPOFA geo-liner specimens after batch equilibrium test and ready for filtration	107
3.23	Duplicated analytes prepared for FAAS analysis	108

3.24	Assemblage of a typical Perspex test cell	115
3.25	Wet-mix kaolin-PPOFA matrix for the compacted liner column	116
3.26	Kaolin-PPOFA geo-liner materials under hydration process	117
3.27	Fabricated laboratory-scale air-pressure system for compressing the geo-mineral liner material in the Perspex column	118
3.28	Compacted kaolin-PPOFA geo-liner column injected with the de-aired distilled water for pozzolanic reaction process	120
3.29	Schematic diagram of the simulated kaolin-PPOFA geo-mineral liner materials connected to the permeameter assemblage	121
3.30	Schematic diagram indicating colour system for the duplicated columns and ease in identification with respect to type of heavy metal aqueous solution used as permeant	125
4.1	Dry state condition of the pulverized palm oil fuel ash	136
4.2	Dry state condition of the industrial kaolin	137
4.3	Variation in the constituted particle sizes of the PPOFA	138
4.4	Variation in the constituted particle sizes of the Industrial kaolin	139
4.5	Loss on ignition and corresponding unburn carbon content of the PPOFA and the industrial kaolin	143
4.6	Surface scanned morphology of the PPOFA magnified at 300 times magnification	145
4.7	Surface scanned morphology of the industrial kaolin magnified at 200 times magnification	146
4.8	Mineralogical composition of the PPOFA	152

4.9	Mineralogical composition of the industrial kaolin	154
4.10	pH and EC of the PPOFA and the industrial kaolin in de-aired distilled water	156
4.11	Estimated liquid limit at 20 mm cone penetration for the industrial kaolin	158
4.12	Compaction curves characteristics for the industrial kaolin modified with PPOFA at varying percentage dose	160
4.13	Relationship between the maximum dry density and optimum moisture content of the four kaolin-PPOFA trial-mixes as a function of percentage PPOFA dose	165
4.14	Dry mixture of 375 g PPOFA in 2500 g kaolin as the formulated geo-mineral	167
4.15	Variation in the constituted particle sizes of the formulated geo-liner material	169
4.16	Surface scanned morphology of the formulated geo-mineral liner material magnified at 200 time magnification	172
4.17	Mineralogical composition of the formulated geo-mineral liner material	176
4.18	Estimated liquid limit at 20 mm cone penetration for the formulated geo-mineral liner	179
4.19	Metal removal efficiency of the formulated geo-mineral liner material	184
4.20	Zn <sup>2+</sup> experimental and isotherm fitness isotherm curves	199
4.21	Pb <sup>2+</sup> experimental and isotherm fitness isotherm curves	199
4.22	Cd <sup>2+</sup> experimental and isotherm fitness isotherm curves	199
4.23	Cu <sup>2+</sup> experimental and isotherm fitness isotherm curves	200
4.24	Hydraulic conductivity of the eight compacted	

	kaolin-PPOFA geo-mineral liner columns	208
4.25	Variation of $C_{\text{eff}}$ , $pH_{\text{eff}}$ , and $EC_{\text{eff}}$ with PV of 0.0001-M $Zn^{2+}$ ions	216
4.26	Variation of $C_{\text{eff}}$ , $pH_{\text{eff}}$ , and $EC_{\text{eff}}$ with PV of 0.0001-M $Pb^{2+}$ ions	217
4.27	Variation of $C_{\text{eff}}$ , $pH_{\text{eff}}$ , and $EC_{\text{eff}}$ with PV of 0.0001-M $Cd^{2+}$ ions	217
4.28	Variation of $C_{\text{eff}}$ , $pH_{\text{eff}}$ , and $EC_{\text{eff}}$ with PV of 0.0001-M $Cu^{2+}$ ions	217

**LIST OF ABBREVIATIONS**

<i>AR</i>	-	Analytical reagent
<i>BAET</i>	-	Batch adsorption equilibrium test
<i>BET</i>	-	Brunauer Emmet Teller
<i>BS</i>	-	British Standard
<i>BSCS</i>	-	British Soil Classification System
<i>BSI</i>	-	British Standard Institution
<i>BTC</i>	-	Breakthrough curve
<i>CCL</i>	-	Compacted clay liner
<i>CE</i>	-	Cation Exchange
<i>CEC</i>	-	Cation Exchange Capacity
<i>CSH</i>	-	Calcium Silicate Hydrate
<i>DDL</i>	-	Diffused double layer
<i>DIW</i>	-	Deionized Water
<i>DW</i>	-	Distilled water
<i>EC</i>	-	Electrical conductance
<i>EDS</i>	-	Energy dispersive spectroscopy
<i>EDX</i>	-	Energy Dispersive X-Ray Analyzer
<i>EDXRF</i>	-	Energy Dispersive X-Ray Fluorescence spectrometer
<i>FAAS</i>	-	Flame Atomic Absorption Spectrophotometer
<i>GHG</i>	-	Greenhouse gases
<i>GPOFA</i>	-	Ground POFA
<i>GRG</i>	-	Generalized Reduced Gradient
<i>HDTMA</i>	-	Hexadecyltrimethylammomium
<i>HFC</i>	-	Hydrofluorocarbons
<i>ICDD</i>	-	International Centre for Diffraction Data

<i>LA</i>	-	Los Angeles
<i>LOI</i>	-	Loss on ignition
<i>MH</i>	-	High plasticity silt
<i>MSE</i>	-	Mean square error
<i>OFAT</i>	-	One-factor-at-a time
<i>OPC</i>	-	Ordinary Portland cement
<i>PKS</i>	-	Palm kernel shell
<i>PMF</i>	-	Palm mesocarp fiber
<i>POFA</i>	-	Palm oil fuel ash
<i>PPOFA</i>	-	Pulverized palm oil fuel ash
<i>PSA</i>	-	Particle size analyser
<i>PV</i>	-	Pore volume
<i>pzc</i>	-	Point of zero charge
<i>SA</i>	-	Surface area
<i>SEM</i>	-	Scanning electron-microscope
<i>SSE</i>	-	Sum of squares of the error
<i>SWGM</i>	-	Solid waste management
<i>TPPOFA</i>	-	Treated PPOFA
<i>W/B</i>	-	Water-binder ratio
<i>XRD</i>	-	X-Ray Diffractometer
<i>ZAV</i>	-	Zero air void

## LIST OF SYMBOLS

$a$	-	Cross sectional area of manometer tube ( $\text{cm}^2$ )
$A$	-	Cross sectional area of compacted liner sample ( $\text{cm}^2$ )
$C_c$	-	Coefficient of curvature
$C_e$	-	Solute concentration in solution at equilibrium ( $\text{mgL}^{-1}$ )
$C_u$	-	Coefficient of uniformity
$C_{eff}$	-	Effluent concentration ( $\text{mgL}^{-1}$ )
$C_{inf}$	-	Influent concentration ( $\text{mgL}^{-1}$ )
$C_o$	-	Initial concentration ( $\text{mgL}^{-1}$ )
$C_R$	-	Relative compaction (%)
$CEC$	-	Cation Exchange Capacity ( $\text{cmol}(+)/\text{kg} = \text{meq}/100\text{g}$ )
$d_{50}$	-	Mean particle diameter ( $\mu\text{m}$ )
$D^*$	-	Effective diffusion coefficient ( $\text{cm}^2\text{s}^{-1}$ )
$D_h$	-	Hydrodynamic dispersion coefficient ( $\text{cm}^2\text{s}^{-1}$ )
$D_L$	-	Longitudinal hydrodynamic dispersion coefficient ( $\text{cm}^2\text{s}^{-1}$ )
$D_m$	-	Mechanical dispersion coefficient ( $\text{cm}^2\text{s}^{-1}$ )
$e$	-	Void ration (dimensionless)
$EC$	-	Electrical conductance ( $\text{mScm}^{-1}$ )
$EC_{eff}$	-	effluent electric conductance ( $\text{mScm}^{-1}$ )
$EC_{inf}$	-	Influent electric conductance ( $\text{mScm}^{-1}$ )
$EC_o$	-	Initial electrical conductance ( $\text{mScm}^{-1}$ )
$EC_e$	-	Equilibrium electrical conductance ( $\text{mScm}^{-1}$ )
$EDS$	-	Energy Dispersive Spectroscopy ( $2\theta$ )
$EDX$	-	Energy Dispersive X-ray ( $2\theta$ )

$f_{oc}$	-	Organic carbon (%)
$f_{om}$	-	Organic matter (%)
$F_d$	-	Diffusive flux ( $\text{mgcm}^{-2}\text{sec}^{-1}$ )
$F_M$	-	Dispersive flux ( $\text{mgcm}^{-2}\text{sec}^{-1}$ )
$F_T$	-	Total mass flux ( $\text{mgcm}^{-2}\text{sec}^{-1}$ )
$F_v$	-	Advective flux ( $\text{mgcm}^{-2}\text{sec}^{-1}$ )
$H_1$	-	Height from effluent point to the upper mark (cm)
$H_2$	-	Height from effluent point to the lower mark (cm)
$H_3^*$	-	Distance of third calibration point from the upper mark (cm)
$H_i$	-	Initial head of the de-aired DW in the manometer (cm)
$H_f$	-	Subsequent head of the de-aired DW estimated in the Manometer at the end of a particular period of time (cm)
$i$	-	Hydraulic gradient (dimensionless)
$I_p$	-	Plasticity index (%)
$k$	-	1-D hydraulic conductivity ( $\text{cms}^{-1}$ )
$k_{ave}$	-	Average hydraulic conductivity ( $\text{cms}^{-1}$ )
$k_{max}$	-	Maximum recorded hydraulic conductivity ( $\text{cms}^{-1}$ )
$K_F$	-	Freundlich isotherm coefficient
$l$	-	Length of sample under test (cm)
$LOI$	-	Loss on ignition (%)
$n$	-	Porosity (dimensionless)
$M_s$	-	Mass of dry liner material add to the aqueous solution (g)
$N$	-	Freundlich isotherm constant (dimensionless)
$pH_e$	-	Equilibrium pH
$pH_{eff}$	-	Effluent pH
$pH_{inf}$	-	Influent pH
$pH_o$	-	Initial pH
$Pe_L$	-	Peclet number (Dimensionless)
$PV$	-	Pore volume ( $\text{cm}^3$ )
$q_e'$	-	Solute removal efficiency (%)



$q_e$	-	Mass of contaminant sorbed per unit dry mass of the solid ( $\text{mgg}^{-1}$ )
$q_{\text{expt}}$	-	Measured mass of contaminant sorbed per unit dry mass of the solid ( $\text{mgg}^{-1}$ )
$q_{\text{eFred.}}$	-	Freundlich mass of contaminant sorbed per unit dry mass of the solid ( $\text{mgg}^{-1}$ )
$q_{\text{eLang.}}$	-	Langmuir mass of contaminant sorbed per unit dry mass of the solid ( $\text{mgg}^{-1}$ )
$r$	-	Linear correlation coefficients
$R^2$	-	Coefficient of determination
$R_d$	-	Retardation factor (dimensionless)
SA	-	Surface area ( $\text{m}^2\text{g}^{-1}$ )
$t$	-	Solute transport time in the porous medium (sec)
$t_i$	-	Start time of test (sec)
$t_f$	-	Subsequent estimated time corresponding to $H_f$ (sec)
$\Delta t$	-	Difference between $t_f$ and $t_i$ (sec)
$T_R$	-	Dimensionless time factor
$V_w$	-	Volume of solution containing an initial concentration, $C_o$ (L)
$V_s$	-	Seepage or average linear velocity ( $\text{cms}^{-1}$ )
$w_L$	-	Liquid limit (%)
$w_{\text{opt}}$	-	Optimum water content (%)
$w_p$	-	Plastic limit (%)
$W$	-	Mass of adsorbent (g)
$XRD$	-	X-Ray Diffraction (count per sec / cps)
$Z$	-	Distance in the direction of contaminant flow (cm)

## LIST OF APPENDICES

APPENDIX	TITLE	PAGE
A	BSCS Sub-group Classification of Soils based on the Grading and Plasticity Chart (BS 5930:1999)	243
B	Properties of Aqueous Solutions Prepared for each Metal Solute	244
C	Typical AAS Results for the Sequential Batch-test at 1:20 mix Ratio with Traces of Pb <sup>2+</sup> detected	246
D	Pore Volume Analysis of the Compacted Kaolin-PPOFA Geo-liner Material	247
E	Minimum Geotechnical Requirements Recommended for Compacted Mineral Liner to Attain Hydraulic Conductivity of $k$ less than or equal to $1 \times 10^{-7} \text{ cms}^{-1}$	251
F	The Calibration Curve obtained for the SPB32-120 $\pm$ 0.001g Analytical Bench-top Balance	252
G	Water Content Determination at 10% PPOFA Dose	253
H	Typical Data Generated for the Determination of Series of Dry Density at the same 10% PPOFA Dose	254
I	Data Points Generated for the Plotting of the Dry Density-moisture Content Relation Curve and the Zero Air Void Line at 10% PPOFA Dose	255

J	Operating Parameters for the Aqueous Environment at Batch Equilibrium for the Formulated Geo-liner Material	256
K	Trend in the Heavy Metal ions Removal Efficiency by the Formulated Geo-liner Material	260
L	The Feasible Data Points Adopted for Regression Parameters and Equilibrium Sorption Isotherm Model	261
M	Linearized Regression Line used to Determine the Parameters for the Freundlich Isotherm	263
N	Linearized Regression Line used to Determine the Parameters for the Langmuir Isotherm	265
O	Freundlich and Langmuir Isotherm Parameters for $Zn^{2+}$ Sorption onto the Formulated Kaolin-PPOFA Geo-liner Material	267
P	Hydraulic Conductivity and Corresponding Statistical Parameters obtained for each of the Kaolin-PPOFA Composite Liner Columns	269
Q	Typical Calculation for the Determination of Hydraulic Conductivity of the Compacted Kaolin-PPOFA Liner Material	270
R	Solute Breakthrough Process of each Metal Solute	271
S	Determination of $D^*_L$ and $R_d$ Parameters from the Excel Solver Nonlinear Generalized Reduced Gradient (GRG2) Optimization Technique	279
T	List of Related Publications	280

## CHAPTER 1

### INTRODUCTION

#### 1.1 Background of the Problem

In recent years the acceleration in urbanization of many countries worldwide has led to interest in various fields of research. One area is in the solid waste management (SWM) ( Foo *et al.*, 2013; Karagiannidis *et al.*, 2010; Chen *et al.*, 2010; Guo *et al.*, 2010). Solid wastes are generally being generated by human activities. These wastes may be classified as municipal wastes from commercial and residential sources; hazardous wastes from hospitals and radioactive plants; as well as agricultural and industrial wastes. Colossal part of these wastes ends up in landfills as they are characterized by their unhygienic nature. Thus, solid wastes are generally unacceptable to the society.

Usually, open dumping or unlined landfilling of generated solid wastes is characterized by uncontrolled and heterogeneous disposal. Though waste dumping is regarded cheap to manage, it is highly pervasive and environmentally unacceptable to the teaming society. It has also been recognized as one of the contributing factors to unhealthy environment. Hence, open dumping could be classified as unsuitable technique militating against the yearnings and aspirations for green and sustainable environment. As such, nowadays unlined landfilling is being replaced by sanitary landfill. The sanitary landfilling of solid waste may be categorized as land disposal technique, and is commonly known as

“engineered sanitary” landfill systems (Ayomoh, *et al.*, 2008; Manfredi and Christensen, 2009; Abu Amr *et al.*, 2012 ).

Even though engineered landfilling approach has been reported as inappropriate (Chang *et al.*, 2009), to date the technique still represents one of the primary component in integrated and sustainable waste management techniques (Galante *et al.*, 2010; Perkoulidis *et al.*, 2010; Agamuthu and Fauziah 2010). The prime objective of incorporating engineered landfill in integrated waste management system is to significantly limit the dangerous health impact from the generated wastes on the society. More to this is that it enhances sustainable and eco-friendly environment (Rajesh and Viswanadham, 2011).

Generally, disposed solid wastes in landfill undergo both biological and chemical degradation. Both processes take place in the presence of water percolated through solid waste deposited. The final aqueous-waste effluent discharged is referred to as leachates. Although engineered landfill techniques have tremendously reduced the paranoid of groundwater contamination from the generated leachates ( Benbelkacem *et al.*,2010; Foo *et al.*, 2013), the formation and management of the aqueous contaminant still pose major problem, thus demand great attention (Mendoza and Izquierdo, 2008; Kim *et al.*, 2009; Umar *et al.*, 2010). For instance, leachate is prone to be a veritable source of non-biodegradable and toxic heavy metals ions disposition. Heavy metals, such as zinc (Zn), lead (Pb), cadmium (Cd), Nickel (Ni), Mercury (Hg) and copper (Cu) constitute part of these contaminant solutes. As such, they are considered toxic to human health and the environment at large (Chalermyanont *et al.*, 2009; Zhang *et al.*, 2013; Silva *et al.*, 2013).

Judging from the characteristics and age of the waste materials deposited in landfills, chemical ionization of heavy metals may as well be characterized by different level of toxicity (Östman, 2008; Zhang *et al.*, 2013). Also, some portion of the toxic aqueous pollutants may escape transfer to treatment plant and be transported through the liner located somewhere within the shallow unsaturated zone of the subsoil (Zhang *et al.*, 2012). Thereafter, the liquid waste may get in contact with the surrounding environments and renders it contaminated. Principally the surrounding environments include the valuable but infinite groundwater resource. Research has shown that leachate is liable to adversely

transform the environment through the creation of imbalance and inhabitable ecosystem (Foo and Hameed, 2009; Silva *et al.*, 2013).

Besides, further decomposition of some chemical compounds present in the leachate may contribute significantly to the emission of obnoxious odor from within the landfill site. If uncontrolled, such emission may render the surrounding air polluted. The release of heat trapping greenhouse gases (GHG) such as carbon dioxide (CO<sub>2</sub>) and methane (CH<sub>4</sub>) are also witnessed (Zhang *et al.*, 2013; Ahmari and Zhang, 2013). Other non-environmentally friendly gaseous emissions such as ammonia (NH<sub>4</sub>), nitrous oxide (N<sub>2</sub>O) and hydrofluorocarbons (HFCs) are also released from the fill. As obtainable in groundwater pollution, the consequential effects of air pollution include adverse human health related implications such as cancer and air borne diseases. From the aforementioned, further research specifically on sustainable earthen landfill liner material is eminent. With this the adverse environmental impacts of the liquefied waste could be effectively ameliorated and at the same time ensure that the liner keep serving as functional integral part of solid waste management system.

The present situation in landfill liner material research shows that there are concerted efforts specifically geared toward the use of minerals as sustainable landfill liner material (Musso *et al.*, 2010; Koutsopoulou *et al.*, 2010; Quaghebeur *et al.*, 2013). Generally, landfill liners constructed from the use of only one material such as soil is not suitable for use as solid waste repository. It may often not meet the regulatory performance requirement of hydraulic conductivity and related contaminants attenuation (Chalermyanont *et al.*, 2009). Hence, soil liners are predominantly design of compacted composite clay material and formed the lower integral part which directly overlies the natural geologic structure (Fall *et al.*, 2009; Guyonnet *et al.*, 2009; Silva and Almanza, 2009; Lange *et al.*, 2010).

More specific, Fall *et al.* (2009) have shown great interest in the use of minerals as landfill liner material. They took the advantage of the strong negatively charged surface energy carried by soil particles. Hence, reactive mineral liner materials are used as trapping mechanism of the positively charged solute components of the contaminants. Usually, these solutes are often trapped and immobilized while migrating through the pore spaces of

the hydrated compacted clay liner (CCL) (Chalermyanont *et al.*, 2009; Silva and Almanza, 2009; Koutsopoulou *et al.*, 2010). The end result was the effective reduction and mitigation of groundwater pollution due to toxic heavy metal ions leached from the solid waste disposed in landfills.

Furtherance to the use of CCL, innovative researches are being carried out on the potential and viability of amending clayey soils with waste materials. The matrixes formed are often used as an alternative composite mineral liner material. These waste materials include ashes from incinerator plants (Travar *et al.*, 2009) as well as bottom and fly ashes produced as by-product from the burning of coal in power generating plants (Nhan *et al.*, 1997; Kayabali and Bulus, 2000; Sivapullaiah and Baig, 2011) Other materials used as amending agent include secondary waste (Ganjian *et al.*, 2004) and natural clay-shredded tyre mixtures (Al-tabbaa and Aravinthan, 1998; Cokca and Yilmaz, 2004) One promising area of application of solid waste as alternative composite mineral liner material which at present lacks attention is in the incorporation of palm oil fuel ash (POFA) as additive material. POFA is a renewable bio-residue ash and is derived from palm oil milling industry. Judging from its physico-chemical properties and previous applications, POFA may be used as a potential cementitious and bio-sorbent additive in earthen sanitary landfill liner.

Large quantity of POFA waste generations are witnessed in rapidly developing countries like Malaysia, Indonesia, Thailand and Nigeria where varieties of oil palm trees species are being cultivated as cash crops (Subramaniam *et al.*, 2008; Yin *et al.*, 2008; Foo and Hameed, 2009; Patthanaisaranukool *et al.*, 2013; Ohimain and Izah, 2014) The powdery form of the ash as pulverized palm oil fuel ash (PPOFA) alone has been used as bio-sorbent in waste water treatment. The works of Zainudin *et al.* (2005) and Foo and Hameed (2009) are of particular interest in the area of study. Furthermore, the chemical compositions of POFA have shown that it may be used as supplementary cementitious material in concrete (Abdul Awal *et al.*, 2011; Ismail *et al.*, 2011; Kroehong *et al.*, 2011; Jaturapitakkul *et al.*, 2011; Aldahdooh *et al.*, 2013; Yusuf *et al.*, 2014; Aldahdooh *et al.*, 2014) . Recently Amat *et al.* (2013) mixed raw POFA with gypsum and clay as binder to produce fire resistive panels used for internal partitions in buildings. As such, POFA may be incorporated into earthen landfill liner material for dual purposes of pozzolanic and

cementitious material. The new composite material may be used as an alternative and sustainable landfill liner material.

However, extensive practical experiences with the application of industrial generated ash-wastes have confirmed the leaching of toxic heavy metals (Asl *et al.*, 2013; Çoruh *et al.*, 2013; Abdel Rahman *et al.*, 2013; Yao *et al.*, 2013; Houben *et al.*, 2013) Thus, the presence and leachability of toxic heavy metals from the use of POFA as earthen landfill liner material should not be ignored.

The uniqueness of this innovative research is that it has significantly contributed in moving to a new frontier the boundary of knowledge connected to the valuable use of POFA. As such, the research has changed the paradigms in the use of the agro-based waste to a new resource, by premixing kaolin with the PPOFA and use as an alternative composite landfill liner material. The research has critically examined the physical, chemical, and mechanical properties of each material component and the formulated composite kaolin-PPOFA liner material. The performance of the formulated mineral liner was examined in terms of its removal efficiency and sorption capacity of four selected heavy metal ions in single-elemental species ( $Zn^{2+}$ ,  $Pb^{2+}$ ,  $Cd^{2+}$ , and  $Cu^{2+}$ ); the leachability of the selected heavy metals; and the hydraulic conductivity. Finally the transient contaminant transport performance of the aqueous solution formed with respective to each of the single-metal ions was examined.

## **1.2 Statement of the Problem**

Recent studies have shown that municipal solid waste, industrial wastes, hospital wastes and other waste types pose major threat to the environment and globally constitute great challenges (Liamsanguan *et al.*, 2008; Zhen-shan *et al.*, 2009; Ngoc and Schnitzer, 2009; Karagiannidis *et al.*, 2010; Zaman, 2014; Farooqui, 2014; Aslani and Wong, 2014). For instance, in Malaysia about 28,500 tonnes of municipal solid waste (MSW) have been reported to have been directly disposed of into various landfills on daily bases (Agamuthu and Fauziah, 2011). The authors reported that most of the landfills in Malaysia contaminate



the environment with the generated leachates as well as free emission of noxious landfill gases to surroundings. Similarly, a survey by Karagiannidis *et al.* (2010) revealed that on yearly bases in Greece, a significant part of the over 14,000 tonnes of infectious solid hospital waste generated are been mismanaged through dumping into household open-disposal sites and landfills after sterilization. In the recent years, China has been reported as the largest developing country in the world. The Landfill disposal technique has being the dominant disposal method for MSW generated in the country. Interestingly, as at 2006, China has 342 cost-effective municipal solid waste landfill sites which functionally accept heterogeneous wastes without requirement for separation (Chen *et al.*, 2010).

Due to its economic advantages and contributions toward the achievement of green and sustainable environment, sanitary landfilling of solid wastes continue to be the most economically viable and final solid waste disposal method (Chen *et al.*, 2010; Perkoulidis *et al.*, 2010; Di Maria *et al.*, 2013; Yang *et al.*, 2013). However, waste containment in landfill sites enhances the concentrations of different dissolved and suspended pollutants as well as non-bio-degradable heavy metal ions leached from hydrated disposed wastes. The leachate formed is injurious to the public health and the environment at large. Landfill leachate as contaminated liquid effluent may percolate through liner material after the decomposition of deposited wastes. The liquid has been recognized as one of the most critical and detrimental issue in landfill operation (Ziyang *et al.*, 2009; Zhan *et al.*, 2013; Li *et al.*, 2013; Gallego *et al.*, 2014).

Despite the reliance of many communities on groundwater as main source of portable water and for culinary consumptions, the level of pervasiveness of its contamination and threat to the public health due to the varieties of toxic heavy metals ions is becoming unbearable (Chalermyanont *et al.*, 2009). Some of the common toxic heavy metals largely present in the leachate include, but not limited to lead (Pb), cadmium (Cd), copper (Cu), zinc (Zn), iron (Fe), nickel (Ni), and mercury (Hg). Consequently, many people have suffered great deal from carcinogenic and water borne ailment in connection with the consumption of leachate contaminated water (Zhang *et al.*, 2009; Nadaroglu *et al.*, 2010; Naser, 2013; Devic *et al.*, 2013).

More so, leachate contaminated groundwater has led to the imbalance in eco-system (Foo and Hameed, 2009). In addition, the liquid contaminant has contributed significantly to ozone layer depletion and greenhouse gas effect through the release of the heat trapping greenhouse gases (GHG) such as carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) (Oonk *et al.*, 2013; Ahmari and Zhang, 2013). Other non-environmentally friendly gaseous emissions as ammonia NH<sub>4</sub> and hydrofluorocarbons (HFCs) are also released to the surrounding environment. Accordingly, the installation of engineered and sustainable landfill liner is imperative since it constitutes one of the most important sub-system in the modern integrated and holistic solid waste management system (Edil, 2003; Demesouka *et al.*, 2013; Di Maria *et al.*, 2013).

The performance of engineered compacted clay liner has been evaluated hydraulically and most importantly on the basis of attenuation of various toxic chemical species (Fall *et al.*, 2009; Sunil *et al.*, 2009; Koutsopoulou *et al.*, 2010; Hamdi and Srasra, 2013; Zhan *et al.*, 2013). Also, the sustainability in geo-liner material formulation will necessitate the introduction and use of locally available solid waste material as additional but relatively cheap and reactive cementitious material. Base on the foregoing, the research explored the potential of compacted kaolin modified with pre-determined dry mass of PPOFA (organic ash) as an alternative and sustainable landfill liner material.

The industrial kaolin used as the main landfill mineral liner was procured in Malaysia. Kaolin is one of the sizeable natural resources deposit in Malaysia (Ariffin *et al.*, 2008). It has been reported that Malaysia has some 112 million tonnes of kaolin reserves located in some of the states (Lee and Teoh, 1992). Thus, indicate high natural deposit coupled with active mining activity (Liew *et al.*, 2012; and Ismail *et al.*, 2013). It has also been reported that the surface chemistry of kaolinitic clay is less chemically reactive (Vizcayno *et al.* 2009). The soil may then be classified as unfavourable for use solely as earthen landfill liner. Despite this, existing literatures have shown that researches have focused on the use of kaolin as geo-liner (Srivastava *et al.*, 2005 and Mockovčiaková *et al.*, 2008). Thus, this research explored the advantage of the inherent chemical compositions of PPOFA and was used in enhancing the less reactive kaolin as alternative composite landfill liner material.

### **1.3 Aim and Objectives of the Research**

The aim of this research is to experimentally study the potential of kaolin admixed with PPOFA as CCL. The specific objectives of the study are:

- i. To establish the physical, chemical and mechanical characteristics of the industrial white kaolin, PPOFA and the formulated composite landfill liner material.
- ii. To determine the attenuation efficiency and sorption capacity of four randomly selected single-element species (Pb, Cd, Zn, and Cu) sorbed onto the formulated mineral liner material via batch adsorption equilibrium test (BAET) and validate the experimental results with the Freundlich and Langmuir model and respective sorption isotherm curves.
- iii. To determine the leachability of the industrial white kaolin, PPOFA and the formulated composite landfill liner material via batch leaching procedure (BLP).
- iv. To determine the hydraulic performance of the compacted kaolin-PPOFA mineral liner material and compare with existing criteria for clay liner.
- v. To determine the diffusion-retardation transport parameters governing the migration of the aqueous solutions of the four single-element species when permeated through the compacted column of the kaolin-PPOFA liner material.

### **1.4 Scope of the Research**

The research was limited to laboratory based experiments, using the treated POFA as admixture to the kaolin. Both materials were obtained within Johor State of Malaysia. Initially the raw POFA was beneficiated by sieving and later subjected to two stages of milling process (i.e., hammer and ball milling). The industrial white kaolin was used as received with no further treatment. The simulated composite mineral liner material was formulated by introducing 375 g of the PPOFA to 2500 g dry mass of the industrial kaolin. Series of preliminary tests were conducted to establish the physical, chemical, and mechanical properties of the two basic component materials and their mixture as the composite liner material. The microstructural analyses conducted include the mass loss on

ignition (*LOI*) analysis at predefined furnace temperature of  $440 \pm 25$  °C and surface scanned morphology analysis using Energy Dispersive X-Ray Analyzer (EDX). Other microstructural analyses performed are the elemental and chemical compositions determined via Energy Dispersive X-Ray Fluorescence (EDXRF) spectrometer as well as the mineralogical composition which was determined via Brucker D8 Advanced X-Ray Diffractometer (XRD). The pH and the electrical conductance (EC) of the suspended solids of the three classes of materials were also determined.

The performance of the composite kaolin-PPOFA matrix as mineral liner was examined from the following criteria:

- i. Metal sorption efficiency
- ii. Adsorption capacity
- iii. Metal leachability
- iv. Hydraulic conductivity
- v. Diffusion-retardation parameters.

Four different toxic heavy metal ion solutions which formed part of those commonly found in landfill leachates and are of significant interest to the environment (i.e., Zn, Pb, Cd and Cu), were prepared as single-metal solution. Subsequently, the respective solutions at varying concentrations were engaged in the determination of the sorption efficiency and capacity of the liner material using the BAET isotherm analysis. The initial concentration ( $C_0$ ) of the four heavy metals used for the metal retention efficiency and isotherm sorption tests ranged from  $10 \text{ mgL}^{-1}$  to  $120 \text{ mgL}^{-1}$ . The presence and leachability of the four heavy metals in the kaolin, PPOFA, and the formulated liner material were examined through the BLP leaching test.

The falling head hydraulic conductivity test was conducted on series of compacted liner specimens prepared from the kaolin-PPOFA matrixes. For the test, eight specimens were prepared and compacted in respective transparent Perspex columns; where de-aired distilled (DI) water was used as the permeant. The completion of the hydraulic conductivity test was immediately followed by the column diffusion-retardation tests. The column test was performed with the influent concentration ( $C_{inf}$ ) of each metal compound maintained at

0.0001M. The generated series of data from the test categories were analyzed and results obtained were compared with existing related literatures. In particular, performance of the formulated kaolin-PPOFA matrix as composite liner was as well compared with standard performance criteria for earthen liner material.

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