LEACHABILITY OF HEAVY METALS FROM CEMENT MORTAR BRICKS MODIFIED WITH WATER TREATMENT ALUM SLUDGE

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A thesis submitted in fulfillment of the requirements for the award of the degree of Master of Science (Chemistry)

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I dedicate this thesis to my beloved family:

My dearest parents, Mr. Paramalinggam & Mrs. Thingalalaky

Jai Sri Hanuman

Mr. Khartigesan & Mrs. Indrani

Mr. Thiyagarajan & Mrs. Gayathry

Mr. Gunalan & Mrs. Kalimah

Mr. Partiban & Mrs. Visalatshi

Sharenya

Reshikha

Thiran

Thebaan

Devesht

Shashinie

My loved one & all my friends.....

My utmost and heartfelt thank you for your love and support

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ABSTRACT

Leachability profiles of aluminium (Al), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), nickel (Ni), lead (Pb) and zinc (Zn) from raw drinking water treatment sludge (WTS), laboratory produced sludge and WTS-cement solidified bricks (CMWTS) were studied to determine the potential of reusing WTS in brick manufacturing. The leachability of the heavy metals was investigated using the extraction method. Leach Tests were performed on WTS obtained from the Semanggar Water Treatment Plant, Kota Tinggi, Johore, Malaysia; laboratory produced sludge, and CMWTS produced using the solidification-stabilization (S/S) technique. Structural identity, chemical composition, effectiveness of the solidification-stabilization (S/S) technique and strengths of bricks were also investigated. Surface and other physicochemical properties were studied using FESEM, SEM, BET-surface area analyzer, XRD, FTIR, TOC, compressive strength test and TG analyzer. Leach tests showed that some heavy metals were leached out from samples in acidic solution but very low levels of heavy metals were leached in water and basic conditions and indicating that the WTS was safe for reuse. When the WTS was solidified in cement mortar, the compressive strength of the bricks increased with increasing curing time, pH of the curing solution and amount of WTS added. However, a reduction of compressive strength was observed at 20% WTS in the CMWTS bricks. It can be concluded that WTS has the potential for reuse in brick manufacturing as the addition of up to 20% WTS in cement mortar produced bricks with good strength properties as well as reduced leachability of the selected heavy metals from WTS.

ABSTRAK

Profil keterlarutlesapan aluminium (Al), cadmium (Cd), kromium (Cr), kuprum (Cu), ferum (Fe), mangan (Mn), nikel (Ni), plumbum (Pb) dan zink (Zn) dari enapcemar mentah rawatan air (WTS), enapcemar yang dibuat di makmal dan bata WTS-pemejalan simen (CMWTS) telah dikaji untuk menentukan potensi penggunaan semula WTS dalam pembuatan bata. Keterlarutlesapan logam berat dikaji menggunakan kaedah pengekstrakan. Ujian larut lesap dilakukan terhadap WTS yang diperolehi dari Loji Rawatan Air Semanggar, Kota Tinggi, Johor, Malaysia; enapcemar yang dibuat di Makmal dan CMWTS yang dibuat menggunakan teknik penstabilan-pemejalan (S/S). Identiti struktur, komposisi kimia, keberkesanan teknik penstabilan-pemejalan (S/S) dan kekuatan bata juga dikaji. Sifat permukaan dan sifat fizikokimia dikaji menggunakan kaedah FESEM, SEM, BET-penganalisis luas permukaan, XRD, FTIR, TOC, ujian kekuatan mampatan dan analisis TG. Ujian larut lesap menggunakan pelarut berasid menunjukan tahap larut resap yang sederhana bagi kebanyakan logam berat. Manakala bagi pelarut air dan keadaan beralkali menunjukkan tahap larut lesap logam berat yang rendah, menandakan bahawa WTS adalah selamat untuk diguna semula. Apabila WTS dipejalkan dengan mortar simen, kekuatan mampatan bata bertambah dengan pertambahan masa, pH larutan pengawet dan amaun WTS yang dicampurkan. Walau bagaimanapun, penurunan kekuatan mampatan berlaku pada 20% WTS di dalam bata CMWTS. Sebagai rumusan, WTS berpotensi diguna semula dalam pembuatan bata kerana penambahan hingga 20% WTS di dalam mortar simen menghasilkan bata dengan sifat kekuatan yang baik dan dapat mengurangkan keterlarutlesapan logam- logam tersebut dari WTS.

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LIST OF ABBREVIATIONS

AAlPS	-	Artificial alum- PAC sludge
AAlS	-	Artificial alum sludge
Al	-	Aluminium
$Al (NO_3)_3.9H_2O$	-	Aluminium nitrate
$Al_2 (SO_4)_3.18H_2O$	-	Aluminium sulphate
APS	-	Artificial PAC sludge
ASTM	-	American society for testing and materials extraction
ATS	-	Artificial water treatment sludge
Cd	-	Cadmium
$Cd (NO_3)_2 .4H_2O$	-	Cadmium nitrate
CH_3COOH	-	Acetic acid
Cr	-	Chromium
$Cr (NO_3)_3.9H_2O$	-	Chromium (III) nitrate
CMWTS	-	Cement mortar-water treatment sludge
Cu	-	Copper
$Cu (NO_3)_2 .3H_2O$	-	Copper (II) nitrate
DDDW	-	Double distill deionized water
DOE	-	Department of environment
EPX	-	Extraction procedure toxicity
FAAS	-	Flame atomic absorption spectrophotometer
Fe	-	Iron
$Fe (NO_3)_3 .9H_2O$	-	Iron (III) nitrate

FESEM	-	Field emission scanning electron microscope
FT-IR	-	Fourier transform infrared spectroscopy
HCl	-	Hydrochloric acid
ICP-MS	-	Inductively coupled plasma-mass spectrometry
MEP	-	Multiple extraction procedure
Mn	-	Manganese
$Mn(NO_3)_2.4H_2O$	-	Manganese (II) nitrate
N_2	-	Nitrogen
NaOH	-	Sodium hydroxide
ND	-	Not detectable
NH_4OH	-	Ammonium hydroxide
Ni	-	Nickel
$Ni(NO_3)_2.6H_2O$	-	Nickel (II) nitrate
PAC	-	Polyaluminium chloride
Pb	-	Lead
$Pb_3(NO_3)_2$	-	Lead (II) nitrate
OPC	-	Ordinary Portland cement
S/S	-	Stabilization/solidification
SAJ	-	Syarikat Air Johor
S_{BET}	-	Branauer-Emmet-Teller surface area
SEM	-	Scanning electron microscope
SPLP	-	Synthetic precipitation leaching procedure
SW	-	Scheduled waste
TCLP	-	Toxicity characteristic leaching procedure
TGA	-	Thermal gravimetric analysis
TOC	-	Total organic carbon

USEPA	-	United States environmental protection agency
WET	-	Waste extraction test
WTS	-	Water treatment sludge
XRD	-	X-ray diffraction
Zn	-	Zinc
$Zn (NO_3)_2 .6H_2O$	-	Zinc nitrate

LIST OF SYMBOLS

mg /L	-	Milligrams per litre
M	-	Molar
ppm	-	Parts per million
ρ_b	-	Bulk density
ε	-	Porosity
ρ_s	-	Particle density

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CHAPTER 1

INTRODUCTION

1.1 General Introduction

In the water industry, raw water is purified by three main processes; coagulation, flocculation and sedimentation. Coagulation is a process of removing dirt and other particles suspended in water (Chu, 1999). Chemicals known as coagulating agents or coagulants are added to water to form particles known as 'flocs' (Ikmalzatul Abdullah, 2009). The flocs are able to attract dirt and small particles present in the water forming large particles that are much heavier than water. Alum is a commonly used coagulant in water treatment and purification (Qaiyum *et al.*, 2011). It is a salt consisting of an alkali metal (such as Na or K) and a trivalent metal (such as Al, Fe or Cr) (Aziz *et al*., 2006 and Greenwood and Earnshaw, 1997). Besides alum, poly aluminium chloride (PAC) is also used as a flocculating agent. PAC is very soluble in water and has a strong adsorptive affinity (Ghafari *et al*., 2009 and Rebhun *et al*., 2000). During sedimentation, the heavy coagulated particles sink and settle to the bottom of the sedimentation tank.

Water treatment sludge (WTS), also referred to by various names including 'water treatment residual' (Verlicchi and Masotti, 2001), 'drinking water sludge' (Zamora *et al*., 2008), 'waterworks sludge' (Hovsepyan and Bonzongo, 2009), and 'alum-derived water treatment sludge' (Zhou and Haynes, 2010) is the solid produced together with the processed drinking water in a typical water treatment facility**.** Figure 1.1 shows typical steps of WTS generation and treatment in a water treatment plant. The sludge is considered a waste material of water treatment.

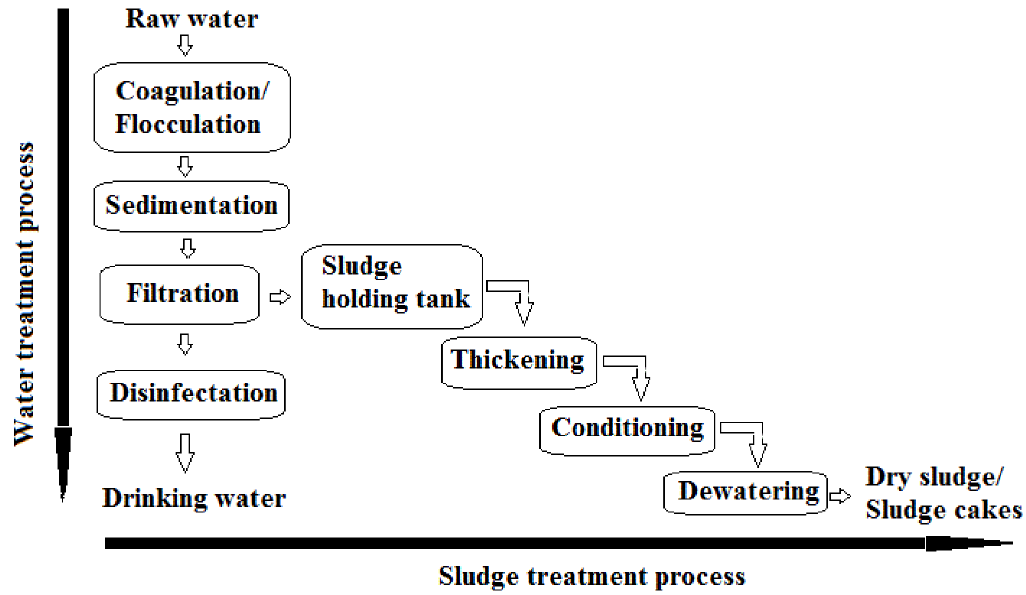

Figure 1.1: Generation and treatment steps for WTS in a water treatment plant.

Alum derived WTS is the most widely generated WTS worldwide, prompting increased concerns with regard to its disposal and beneficial reuse (Zhou and Haynes, 2010 and Verlicchi and Masotti, 2001). Reviews on options for reusing or recycling WTS had been documented, such as the use of WTS as adsorbents in wastewater treatment and as construction materials in constructed wetlands (Zhou and Haynes, 2010, Zamora *et al*., 2008 and Babatunde and Zhao, 2007).

In Malaysia, the disposal of WTS is an integral part of the operation and management of water operators (water treatment utilities) due to stringent regulations on waste management by the Department of Environment (DOE, 2005). A study on the characteristics of WTS generated by selected water treatment plants in Malaysia had shown that although WTS contained heavy metals, the levels were lower than the toxicity characteristic leaching procedure (TCLP) limits (Aminudin, 2009). However, there were still some concerns over the issue of long term accumulation of heavy metals in landfills.

The stabilization/solidification (S/S) technique typically involves the mixing of a solid waste material with a specified binder to reduce the leaching of contaminants from the waste material by either physical or chemical means (Al-Tabba and Perera, 2006). S/S helps to convert an initially hazardous material into an environmentally acceptable form. Thus the waste material may be disposed off safely, or it could be used as construction material. S/S has been widely used in the disposal of many types of hazardous wastes, as well as in the remediation of contaminated disposal sites. Cement has been identified as the most widely used material in the S/S technique compared to other materials (Stegemann and Zhou, 2009). Cement based S/S technique is relatively low cost and has shown good and long term stability (Garrabrants and Kosson, 2005). Specific to the S/S technique, leaching characterization can be used to evaluate waste acceptance for disposal or reuse. S/S waste forms are typically subjected to leaching tests in order to predict environmental impact of trace contaminants such as heavy metals.

The goal of leaching evaluation is to determine the potential for toxic constituent release by leaching from a waste matrix under a management scenario. It determines whether the potential constituent release will be affected by alteration of the release conditions or long-term interactions with the release environment. The 'Leach Test' is a method used to classify waste material for disposal options. It quantifies the amount of material such as metal that is leached out from solid compounds.

Different countries or regions of the world apply specific standard leach tests that vary slightly in the leaching procedure. In the UK Leach Test, a solid to leachant ratio of 1 to 10 is exposed to an extraction time of one hour using the orbital shaking technique. The Japan Leach Test is identical to the UK Leach Test, but the length of extraction is six hours (Zaiton Abdul Majid, 2004). The Toxicity Characteristic Leaching Procedure (TCLP), a leach test proposed by the US Environmental Protection Agency (EPA), has a longer extraction period of approximately 18 ± 2 hours with a solid to leachant ratio of 1 to 20, and makes use of a different shaking technique (Perera *et al.*, 2005).

The Semangar Water Treatment Plant is one of the Water Treatment Plants under the supervision of the Syarikat Air Johor (SAJ) Holdings Private Limited. It is a 120-acre water treatment facility located in Kota Tinggi, Johor. The handling capacity is 160 MLD of treated water to serve the demands for clean water in the Johor Bahru District and the surrounding environments. Raw water is pumped from

Sungai Johor at an abstraction point near to Kampong Semangar, approximately 1 km from the treatment plant.

Wet WTS generated by the plant is separated from residual water, and the water is discharged back into Sungai Johor. The discharged water must be in compliance with the standard ‘A’ of the DOE requirements. The WTS generated by the Semangar facility is stored for 1 year to harden and dry in one of three sludge lagoons (capacity of 35,000 m^2 each), operated in rotation. Every 3 years, the lagoon is layered with sand to replenish the thickness of the sand in the base of the lagoon. Dry sludge is excavated and transported to landfills. The WTS is classified as scheduled waste and elimination is regulated by the Environmental Quality Act 1974 (Act 127). The Semangar Plant has a 40-acre landfill area designed to support 30 years of operation for disposal of sludge. The Water Treatment plant accommodates as much as 120 tonnes of raw sludge and generates an estimated 80 tonnes of dried sludge per year.

1.2 Problem Statement

In Malaysia, an estimated over 2.0 million tons of water treatment sludge or residue (WTS) is produced annually by the water operators throughout the country. The sludge produced is categorized as scheduled waste (SW204), as it contains metals and chemicals and has to be disposed off in accordance with World Health Standards (DOE, 2005). Dewatered residues are sent to the scheduled waste facilities at Kualiti Alam Sdn. Bhd., Negeri Sembilan. Disposal of the sludge involves high operating costs for water operators that can lead to the increase in water tariff. In addition, serious implications can occur, such as accumulation of WTS and toxic materials in the landfill facility.

1.3 Objectives of Research

This research was undertaken to determine the potential reuse of WTS generated by the Semangar Water Treatment Facility, Kota Tinggi, Johore, as a construction material. The main aims of this research were to study the properties of WTS for safe disposal in terms of heavy metal leachability, and the reuse of the WTS as construction material by using the solidification-stabilization (S/S) technique. The following are the objectives of this study:

1. To determine the optimum conditions and effectiveness of alum for heavy metals (iron (Fe), manganese (Mn), nickel (Ni), lead (Pb), aluminium (Al), cadmium (Cd), chromium (Cr), copper (Cu), and zinc (Zn)) removal from aqueous metals solutions.

2. To determine the leachibility of heavy metals from alum sludge generated in the laboratory (AAlS), PAC sludge generated in the laboratory (APS), alum-PAC sludge generated in the laboratory (AAlPS) and raw water treatment sludge generated by a waterworks operator (WTS).

3. To investigate the leachability of heavy metals from WTS-cement mortar bricks (CMWTS) where WTS is immobilized in cement mortar using the stabilization-solidification (S/S) technique.

4. To characterize the properties of WTS and CMWTS using techniques such as Fourier transformed infrared spectroscopy (FTIR), X-ray diffraction (XRD), field emission scanning electron microscope (FESEM), scanning electron microscope (SEM), BET surface analyzer, total organic carbon (TOC) analyzer and thermal gravimetric (TG) analysis.

5. To study the capability of reusing the CMWTS as a material for the construction industry.

1.4 Scope of Research

The first part of the study was focused on the coagulation and flocculation of nine heavy metals (Cd, Zn, Pb, Mn, Cu, Ni, Fe, Al, and Cr) from aqueous metal solutions by alum and PAC. This was to simulate the generation of sludge (AAlS, APS and AAlPS) in the laboratory and to assess the capability of alum and PAC to remove heavy metals at various pH from aqueous solutions.

In the second part, the leachability of the selected heavy metals from AAlS, APS, AAlPS and WTS was studied using five eluents (0.1 M hydrochloric acid, 0.1 M acetic acid, distilled water, 0.1 M ammonium hydroxide and 0.1 M sodium hydroxide). The purpose of the study was to determine the profile of heavy metal leaching from the AAlS, APS, AAlPS and the WTS.

The third part of the study involved the characterization of WTS. The heavy metal content of the WTS was determined using acid digestion and flame atomic absorption spectrophotometer (FAAS). Various methods such as Fourier transformed infrared spectroscopy (FTIR), X-ray diffraction (XRD), field emission scanning electron microscope (FESEM), scanning electron microscopy (SEM), total organic carbon (TOC) analyzer, BET surface analyzer and thermogravimetric (TG) analysis were used to characterize the physicochemical properties of WTS, cement mortar (CM) and cement mortar-water treatment sludge (CMWTS) bricks. The bulk density, particle density, porosity, particle size and pH of the WTS were also analyzed.

Lastly, the WTS was incorporated into cement mortar using the S/S technique to assess its potential for usage as a construction material. CMWTS bricks were constructed and studied in terms of structure, leachability in various solutions and compressive strength.

1.5 Significance of the Research

The results of this research will give an account of the leachibility characteristics of heavy metals bound to alum sludge. The data can determine whether the alum sludge sample from the water operator is safe for disposal at a dumping ground or is suitable for reused as a construction material, or otherwise be categorized as scheduled waste. Disposal of the sludge at ordinary dumping grounds can help water operators reduce cost, and in turn lower water tariff. In addition to the potential and beneficial reuse of the sludge waste for construction, the results can help to educate water works operators, municipalities and the general public on the benefits of the WTS environmental usage.

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