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Cost-Effective Re-Use of Aluminium Based Water Treatment Residuals (Al-WTRs) in Wastewater Treatment

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Abstract- **In typical drinking water treatment plant, Aluminium based-Water Treatment Residuals (Al-WTRs) are generated from coagulation-separation process, in which Fe or Al salts are added to raw water for the removal of colloids, color, suspended solids and common contaminates. The common disposal options of water treatment residuals includes directly discharging to surface waters, discharge to wastewater treatment plants, landfilling and land application. In the letter case, treatment and transportation of the dewatered residuals to a landfill and landfill tipping fees can represent a significant cost to the utility and technical problems. The re-use of Al-WTRs not only provides a low cost technological solution in wastewater treatment process, but also an effective residuals management option for water treatment facilities. This is because, the Al-WTRs have a high content of aluminium hydroxides become the dominant constituent and it also act as a biofilm carrier, making it possible raw material in wastewater treatment that removes various pollutants such as phosphorus, nitrogen, suspended solids, COD and BOD. The beneficial utilization of Al-WTRs in wastewater treatment process, thus converting the Al-WTRs as a useful material, rather than a waste for landfill. Obviously, it is an attempt to use 'waste' for wastewater treatment, demonstrating a 'win-win' technique.**

Index Terms- **Aluminium based Water Treatment Residuals (Al-WTRs), Wastewater, Phosphorus, Nitrogen, Adsorption, Denitrification.**

I. INTRODUCTON

Water treatment processes that are used to produce safe drinking water generate a wide variety of residual products depending on the untreated water source, chemicals used for purification, and types of unit operations used. In the conventional coagulation-filtration treatment process, suspended solids and natural organic matter are removed from the raw water supply by the addition of aluminum and iron salts as coagulants, resulting in the production of water treatment residuals (WTRs) **[1].** The high production of WTRs over the world, treatment and disposal induces cost and technical problems. The reuse of WTRs not only provides a low-cost technological solution, but also an effective residuals management option for water treatment facilities. Alum sludge is a locally, easily and hugely available material with nature of free of charge. Beneficial reuse of the alum sludge falls into the theme of environmental sustainability which encourages "Reduce, Reuse and Recycle" **[2].** This paper therefore presents a comprehensive review of available literature on attempts at cost-effective reuse of water treatment plant sludge in wastewater treatment process, in an effort to provide a compendium of recent and past developments, and update our current state of knowledge **[3].** Nitrogen (N) and Phosphorus (P) are two essential nutrients for all forms of life on earth. Phosphorus can exist in various phosphate species, which are classified as orthophosphates, condensed phosphates, and organic phosphates. Orthophosphate is of significant concern because it is not only the most abundant form of P in water and wastewater, but also is the form that can be immediately utilized by organisms **[4]**. The excess release of phosphorus into surface water may cause eutrophication, and subsequently, deteriorate the water quality. The critical concentration of P above which the growth of algae and other aqueous plants accelerates, is suggested as 0.01 mg/L for dissolved P and 0.02 mg/L for total P **[5]**. Excessive concentration of NO_3^- intake by human body through drinking water or food can cause methemoglobinemia (blue baby syndrome) or carcinogenic effects. The management of phosphorus (P) and nitrogen (N) in natural waters has long been of importance from environmental, health, and economic perspectives. Because of the requirements of costs, operation and maintenance for removal techniques of nutrients in wastewater treatment plants (WWTPs) are usually not available for small-scale and decentralized treatment systems, such as WWTPs in rural areas or small towns, septic ponds and waste lagoons, which are still being widely used in this country **[4].** Therefore, the development of low-cost and efficient technologies for phosphorus and nitrogen removal in small-scale and dispersed treatment systems can have an important impact on water quality. Aluminum based water treatment residuals (Al-WTRs) have a high content of metal (hydr) oxides and organic carbon that remove various pollutants, especially phosphorus and nitrogen, from wastewater. A novel approach of using Al-WTRs to reduce the P and N from the wastewater, which shows the cost-effective re-use of alum sludge rather than treated as a waste for landfill.

II. WATER TREATMENT RESIDUAL CHARACTERIZATION

The physical, chemical, and mineralogical properties of alum sludge are shown in Table 1. The alum sludge was chemically mainly composed of Si, Al and Fe, and had a relatively high content of loss on ignition (LOI) **[5]**.

Table 1. Physical, chemical and mineralogical properties of alum sludge.

Chemical composition	Dominant mineralogy	Texture	Reactive Al fractionation (mg/kg)
Si 32.5%	Kaolinite	sand 82.1%	Exchangeable 7.6
Al 29.4%	Mica	silt 5.6%	Amorphous 21,100
Fe 6.3%	Quartz	clay 12.3%	Organic 6,620
LOI 26.9%	-	-	-
pH- 7.3	Cation exchange Calcium carbonate Organic C capacity (CEC)- 8.1 cmol/kg	Calcium carbonate equivalent (CCE)- 4.2%	Organic Carbon- 6.4%

(Source: Jae Gon Kim[a], Jung Hyun Kim[b], Hi-Soo Moon[b], Chul-Min Chon[b] and Joo Sung Ahnc, 2003)

Sand is the dominant size fraction of the alum sludge. Most silt and sand particles were identified as aggregates of clay and fine silt particles under scanning electron microscopy (SEM) analysis after the removal of cementing materials such as carbonate, organic matter, and iron oxide (Figure 1) **[5]**.

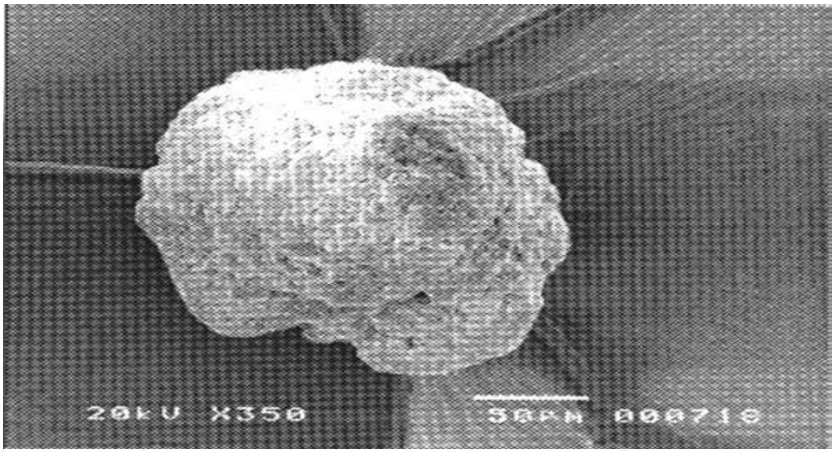

Figure 1 SEM micrograph of sand size aggregates of alum sludge

Water treatment residuals tend to be amorphous in nature. For example, as shown in Eq., when alum is added to water it reacts with bicarbonate to form amorphous $Al(OH)_{3(s)}$: **[1]**

$$Al_2(SO_4)_3{\bullet}14H_2O + 3Ca(HCO_3)_2 \rightarrow 2Al(OH)_{3(s)} + 3CaSO_4 + 6CO_2 + 14H_2O$$

Due to their porosity and amorphous nature and the presence of Al (hydr) oxides, WTR have the propensity to adsorb tremendous quantities of anions. Anion sorption on to WTR should be a function of the WTR particle size, charge, and surface area **[1]**. However, the nature of WTRs makes it possible to act as a biofilm carrier for denitrification. Bearing a high content of an active carbon (C) source, the WTR surface can facilitate the biological denitrification process and have the potential to achieve N removal **[4]**.

III. LITERATURE REVIEW: BATCH EXPERIMENTS

Qian Liang, 2011 used Al-WTR was placed into 250 ml flasks with 100 mL of either WW or SS and the mixtures were shaken on a rotary shaker with a speed of 175 rpm with both the wastewater (WW) and a synthetic solution (SS) prepared with D.I. (deionized) water spiked with KH_2PO_4. The results demonstrated good removal efficiency for both reactive P (RP) and total P (TP) by Al-WTR solid with adequate dosage. When the sorbent dosage reached or exceeded 6 g/L, 95% of RP and 90% of TP could be removed, and equilibrium concentrations of about 0.06 mg RP/L and 0.20 mg TP/L could be achieved **[1]**. Babatunde, A.O.; Zhao, Y.Q.; Burke, A.M.; Morris, M.A.; Hanrahan, J.P.,2009 perform a batch to determine the P adsorption capacity and adsorption parameters. 1.0 g of the Al-WTR was equilibrated with 100 ml of varying P concentrations ranging from 0 to 360 mg-P L^{-1} for 48 hours at different pH levels (pH 4, 7 and 9). The adsorption capacity decreased with an increase in pH from 4 to 9, indicating that the adsorption process is more favoured under acidic conditions. The pH at the point of zero charge (pHpzc) also plays an important role in the adsorption phenomenon. At pH below the pHpzc, the surface would be positively charged. Therefore, at low pH (with abundant positive sites), phosphate adsorption will be facilitated by electrostatic and chemical attraction onto the positively charged surface, but as the pH rises towards and above the pHpzc, the surface becomes predominantly negatively charged due to competitive adsorption of OH- and phosphate adsorption decreases. A maximum adsorption capacity of 31.9 mg-P g^{-1} was obtained at pH 4, about three times the value obtained at pH 9 **[6]**. M. Razali, Y.Q. Zhao* and M. Bruen, 2007 used three types of phosphate suspensions were prepared separately; 14.7 mg-PO_4^{3-}/L orthophosphate (potassium dihydrogen phosphate: Riedel De Haen KH_2PO_4, AnalaR grade), 10.8 mg-PO_4^{3-}/L polyphosphate (sodium hexametaphophate: BDH $(NaPO_3)_6$, no grade) and 3.3 mg-PO_4^{3-}/L organic phosphate (adenosine monophosphate: Fluka ($C_{10}H_{14}N_5O_7P{\cdot}H_2O$), AnalaR grade). Different masses of alum sludge ranging between 0.1g to 0.5 g and 100 ml of P suspension were poured into 150ml plastic bottles and the pH values of the suspensions were adjusted to 4.0, 5.5, 7.0 and 9.0 respectively by adding 0.1M sulphuric acid and 0.01M sodium hydroxide. The mixed samples were placed on a Stuart Orbital Shaker to agitate at 200 rpm for various durations up to 24 hours although different equilibration times of 17 hours, 24 hours, 48 hours, 6 days and up to 80 days with initial P concentration ranged from 5.0 to 3,500 mg P/L. The adsorption capacities vary for each phosphate type where orthophosphate has the highest adsorption capacities for all pH values tested while organic phosphate removal was the slowest. The adsorption capacity for orthophosphate was 10.2 mg-PO_4^{3-}/g sludge, polyphosphate was 7.4 mg-PO_4^{3-}/g sludge and organic phosphate was 4.8 mg-PO_4^{3-}/g sludge, all at pH 4.0. Higher removal efficiencies were recorded at lower pH ranges while lower removal rates were observed at higher pHs **[7]**.

IV. LITERATURE REVIEW: COLUMN EXPERIMENTS

Yaqian Zhao, 2010 used Six different vertical flow CW (VFCW) reactors were constructed using Pyrex columns (1.0 m long, Ø = 0.095 m). The Pyrex columns were filled with the dewatered alum sludge and overlain with either pea gravel or sand in different proportions, ranging from 0 to 60%. The total duration of the experiments was 25 weeks. Reactor 6 was operated in a continuous up-flow mode to examine any effect of the batch feeding on P removal and also any reduction in clogging tendency by avoiding surface filtration of the suspended solids (SS). At the end of the experimental period, an average overall P removal efficiency of 93.1, 91.9, 79.5, 67.1, 89.8 and 86.9% (SRP), and 87.3, 87.8, 77.0, 69.0, 86.4 and 83.6%

(RP) was achieved in Reactors 1, 2, 3, 4, 5 and 6 respectively. Reactor 5 had an average overall removal efficiencies of 66.4% SS, whereas Reactors 1 and 2 had average overall removal efficiencies of 64.6 and 63.0% SS respectively, giving a marginal difference of 1.8 to 3.4% in the removal efficiency **[2]**. Continuous-flow column tests were conducted with filter media packed columns in the laboratory. The filter media used included sand, Al-WTR and 35 mixtures of the two with different proportions. Clear PVC pipes were utilized as the columns, and each of them had a length of 30.5 cm and an inner diameter of 2.5 cm. In order to prevent photosynthesis and algae growth in the filter media, foil was used to cover the columns and shield the light from outside. Simultaneous P and N removal was achieved in column tests with high efficiencies for P (95.99% for RP and 90.04 % for TP) and N (99.62% for nitrate). The maximum sorption densities obtained at pH of 4, 7, and 9 **[4]**. Nazirul mubin ZAHARI*, chua kok HUA, lariyah mohd SIDEK used a cylindrical transparent column was used in this study with the inner diameter of 7.4 cm and 100 cm long. The gravel was placed at the base of the tube to prevent the losses of the dried alum sludge and support the alum sludge. The wastewater sample was collected at the bottom of the column. The pump was used to recycle the wastewater for continuous flow until the end of the experiment. This study was operated with constant flow rate of 5.16 $x10^{-1}$ m^3/d. The initial concentrations of influent and effluent wastewater sample are 6.52 mg/L and 2.96 mg/L respectively. Effectiveness of phosphate removal on river water application: The maximum percentage phosphate removal rate is 75.9%. Effectiveness of phosphate removal on wastewater samples: The maximum removal rates are 95.2% and 89.49% respectively for influent and effluent sample **[8]**. Yang Y.; Zhao, Y.Q.; Babatunde, A.O.; Kearney, P., 2009 used Al-WTR as a potential co-conditioner and dewatering agent in anaerobically digested biosolids. A 2:1 biosolids/Al-WTR ratio was the optimal mix ratio on a volume basis, resulting in 99% P reduction in rejects waste water. Th e authors also showed that Al-WTR enhanced the dewaterability of biosolids because the Al-WTR played a role in charge neutralization and lowered the specific resistance to filtration and capillary suction time. Polymer dosage, for biosolids dewatering purposes, could also be reduced from 120 to 15 mg L^{-1} with Al-WTR usage and should be reflected as a substantial cost savings to municipalities. Reject water filtration with alum sludge bed: The experimental setup consists of a Perspex column (1.0 m long and 94 mm in-diameter) packed with 3.3 kg of prepared dewatered alum sludge to a height of 65 cm with 10 cm of gravel at the base to act as support layer. The reject water, which is a mixture of the supernatant from a gravity belt thickener and the supernatant from a centrifuge, with average P concentration of 113.5mg-P/L was passed through the bed from the top of the bed at a filtration velocity of 1.0 m/h. The second strategy of reject water filtration with alum sludge bed has shown a good performance of P reduction **[9]**. Hu, Y.S.; Zhao, Y.Q.; Zhao, X.H.; Kumar, J.L.G. used plexiglass column (diameter 9.3 cm). 10cm-depth gravel was filled into the bottom as the support medium and 60cm-depth air dried dewatered alum sludge (2 kg, moisture content 74%, particle size 1-3 cm) was filled as the main wetland medium layer, which gives a total volume of 4.75 L with initial porosity of 42% (2 L liquid contained). The system was operated in batch mode with 3 cycles per day. Only 56% NH_4-N removal was obtained on average. It was quickly increased to 85% with effluent NH_4-N of 51 mg/L 16 after the enhancement of air supply in second period. Extreme nitrification was recorded in period 3 with average NH_4-N removal of 97% and effluent NH_4-N of 12 mg/L. Both the COD and BOD_5 degradation were achieved with average removal of 74% and 82%, respectively **[10]**. Babatunde, A.O.; Zhao, Y.Q. showed that Aluminium hydroxide sludge discharged to a sewer in a treatment plant has proved completely successful with phosphate removal up to 94%, at a dose ratio of 0.3 to 1 corresponding to about 3.5m mole/l of Al. Both SS and COD removal efficiencies were improved by 20% and 15% respectively at a sludge dose of 18-20mg al/l. The waterworks sludge could be more effective at reducing potential runoff P loses than its use as an amendment to lower P-concentration **[3]**.

V. CONCLUSIONS

It has been seen from the literature that, an aluminium-based water treatment residual (Al-WTR) from a local water treatment facility can be effective adsorbent for phosphorus (P) and nitrogen (N) removal in wastewater. The processes of phosphorus-sorption and denitrification in the Al-WTRs, as two essential removal mechanisms. Where Al-WTR has a high content of aluminium hydroxide, which is able to bond phosphorus in solution mainly via ligand exchange. Also, Al-WTR is found to be rich in bio-available carbon, which can facilitate biological denitrification for N removal. These are two essential removal mechanisms of phosphorus (P) and nitrogen (N) in wastewater. It has an ideal surface for biofilm growth and attachment with specific surface area.

Removal mechanism of phosphorus (P): Ligand exchange of phosphate ions and metal hydroxides on media surfaces is believed to be the predominant mechanism for the sorption of phosphorus. When contacting with aqueous solutions, the metal (hydr) oxides on the surface of the sorbent material become hydrated or hydroxylated, serving as the binding sites. Phosphate anions can be exchanged with the hydroxyl ions on the solid surface to form surface complexes, thus being immobilized. For orthophosphates the ligand exchange reaction can be described as shown in Equation:

$$aMOH_{(s)} + H_bPO_{4(aq)}^{(b-3)} + cH^{+}_{(aq)} \leftrightarrow M_aH_cPO_{4(s)} + l$$

Where M stands for a metal ion (usually Al of Fe), and b (≤3) refers to the degree of protonation of an orthophosphate ion. Figure 2 shows the possible forms of phosphorus surface complexes.

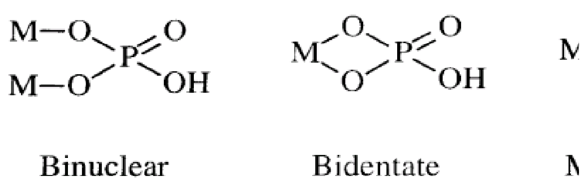

Figure 2 Forms of surface complexes

Removal mechanism of nitrogen (N): The most prevalent forms of nitrogen in wastewaters are organic, ammonium and nitrate nitrogen. However, only less than 1% of N exists in the oxidized form of nitrate in raw wastewater. Typically organic (60%) and ammonium (40%) forms of N are the dominating forms in domestic water. Nitrification and denitrification are two essential processes for biological N removal. Nitrification is the biological oxidation of ammonia (NH_3)/ammonium ($NH^{4+)}$) to nitrate with nitrite as an intermediate. Nitrification depends on the presence of both nitrifying organisms (autotrophic) and oxygen. Denitrification is the biological conversion of nitrate-nitrogen to more reduced forms such as N_2, N_2O and NO. The requirements for denitrification include the denitrifying organisms (autotrophic or heterotrophic), nitrate ions, carbon source and the absence of oxygen. Figure 3 shows Carbon sources for denitrification from different ways.

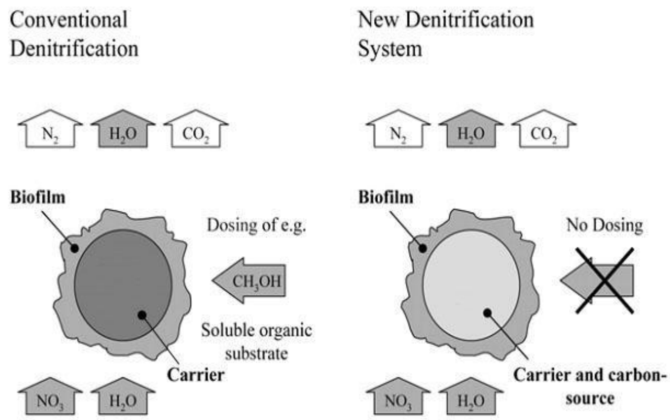

Figure 3 Carbon sources for denitrification from different ways

Al-WTR is found to be rich in bio-available carbon, which can facilitate biological denitrification for N removal. It can achieve better removal efficiency in a low-carbon solution while reducing the demand of a supplemental carbon source. When the carbon source from the solution was available, it is possible that the biofilm in WTR-WW is able to obtain organic carbon (OC) from both inside substrate and outside solution **[4]**.

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