A Two-Prong Approach of Beneficial Reuse of Alum Sludge in Engineered Wetland: First Experience from Ireland

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Abstract Effective management of the industrial waste requires a sustainable approach that maximises its value of reuse/recycle for other industrial demands and the environment needs. This article aims in exploring the potential of the intended purposes in the newly developed dewatered aluminium-water treatment sludge (Al-WTS)-based engineered wetland (EW) for wastewater treatment. Due to the low energy requirement and aesthetical appearance, EW is seen as a ‘green’ wastewater treatment technique worldwide, for a wide variety of wastewater treatment. The Al-WTS-based EW developed at University College Dublin, Ireland, represents the latest initiative at using engineering ingenuity to further improve EWs performance. This article summarises the background of development and the results derived from different phases of the development of Al-WTS-based EW.

Keywords Alum sludge (Al-WTS) · Adsorption · Engineered wetland · Reuse · Wastewater treatment

Alum Sludge and Its Concerns

It is well noted that the generation of coagulant residual sludge in the current potable water treatment technologies may remain unavoidable. Alum sludge (or Al-water treatment sludge i.e., Al-WTS) is generated at water treatment plants worldwide where aluminium sulphate is used as the primary coagulant (Fig. 1).

Although Al-WTS is currently classified as ‘nonhazardous’ by the current EU legislation, the daily production of the increased vast amount is triggering off considerable environmental and economic concerns as well as disposal issues. In Ireland, 18,000 t dry solids of Al-WTS in an annual basis is generated with landfill disposal costs of about €3.2 million. In the UK, about 182,000 t dry solids of waterworks sludge is generated each year, with disposal to landfill as the predominant disposal route. Since the drinking water supply is the number one priority of the human life, the production of Al-WTS as by-product in water treatment plants is continuous along with the human daily life. Accordingly, the disposal of the AL-WTS has become mandatory for water companies especially in recent years due to increasing environmental awareness, escalating costs, dwindling landfill space and the need for sustainability. Thus, the search for cost effective and eco-friendly disposal option(s) of Al-WTS becomes an urgent priority.

Engineered Wetland and Its Needs for Further Development

In Ireland, approximately 82% of Irish urban wastewaters receive secondary treatment. Among the treatment facilities, there are 144 engineered wetlands (EWs) under operation across the country according to a survey [3]. This makes a significant and measurable contribution of EWs to the Irish water environmental control. EW has been well recognised as a ‘green’ wastewater treatment technique worldwide due to its low energy requirement and aesthetical appearance. It has been increasingly applied globally...
for the treatment of various wastewaters [13]. It is noted that the performance of the EWs is generally good in terms of the removal of organics (termed as COD and BOD₅) and suspended solids (SS), but as regards nutrient (N and P) reduction, their performance has been inconsistent and often low [9]. There is even more concern when EWs are employed to treat medium to high strength wastewater with high nutrient (especially phosphorus (P)) concentration. This would require alternative substrates (rather than normally used soil, sand, gravel, crushed stone, etc.) with high P sorption capacity in order to reduce P concentration to acceptable levels. In addition, regarding the treatment of high strength wastewater, there is a need to enhance the oxygen transfer efficiency in the EWs to improve the organics removal. Although vertical sub-surface flow EWs were developed in good configuration to improve the aeration, operational strategy of such kind of EWs may have the space to further promote the oxygen supply/transfer. These are the driving force for the new development of EWs.

**Proposed Reuse of Al-WTS in EW: A Solution with Two-Prong Feature**

A research attempt was made to probe the possible reuse of the Irish dewatered Al-WTS as a low cost alternative substrate in EWs, particularly for P removal in wastewater treatment, towards developing the novel Al-WTS-based EWs system. Since dewatered Al-WTS has the following unique features compared with other industrial by-products, such as slag, fly ash, etc.

- Al-WTS is predominantly composed of amorphous aluminium ions (29.7 ± 13.3% dry weight [1]), it can be used as a valuable material in the treatment of wastewater as the ions enhance adsorption and chemical precipitation processes that remove various pollutants, especially P.
- Al-WTS is an unavoidable by-product in water treatment plants (Fig. 1). In particular, it is easily, locally and hugely available and free of charge for the moment.
- Al-WTS is relatively clean with respect to heavy metals and organics, and poses lower environmental risks compared with other industrial wastes [6].

Therefore, if adopted in EWs, the two-prong approach of significant cost savings on both existing Al-WTS disposal and wastewater treatment via EWs, can be achieved. This is, of course, contributed to sustainable development by using ‘waste’ for wastewater treatment. In the Centre for Water Resource Research, University College Dublin, Ireland, a wetland research group has conducted the extensive study to identify the characteristics and the P adsorption capacity of a local Al-WTS and has demonstrated that the Al-WTS is a reliable and cost-effective material for P-rich wastewater treatment [4, 14]. More significantly, a so-called novel engineered wetland system for high strength P-rich wastewater treatment has been
developed in laboratory scale basis by employing local Al-WTS as main substrate in the EWs [16]. Figure 2 illustrated the road map and the objectives of each phase in such the development. Currently, the development is focusing on the field demonstration study to validate the results and established principle obtained from the laboratory investigations, which have been completed. Overall, adsorption of P by Al-WTS was firstly investigated by batch tests. The maximum adsorption capacity (Q₀, mg/g) was obtained by fitting Langmuir isotherm and the typical result of Q₀ is given in Table 1. It shows that the Al-WTS is a promising low cost adsorbent with excellent high P adsorption ability, which is capable to compare with other industrial by-products [11].

A single model EW employing 100% dewatered AL-WTS as substrate was then set up and operated on short term [15] and a long term basis [16] under a newly developed ‘tidal flow’ operation strategy. Tidal flow is under the principle of artificially creating the ‘tide’ i.e., the rhythmical filling and draining of the EW medium, by peristaltic pump controlled by timer [7, 12]. It has been demonstrated that the tidal flow operation can enhance the oxygen transfer in EW, and thus make it possible to treat relatively high strength wastewater. Following the success of trial of the single Al-WTS-based engineered wetland, a multi-stage EW system was designed and operated to treat an animal farm wastewater under various organic loadings and operating conditions. The study was also focused on addressing the lifetime of the Al-WTS-based EW system and the final disposal of the Al-WTS when it becomes fully saturated in the EW. The main findings of the laboratory scale development of the Al-WTS-based EW are summarized in Table 2. The high removal efficiencies on COD, BOD₅, P, SS showing in Table 2 provides the ample evidence that the Al-WTS can be employed as substrate to serve as filter medium for SS removal and as biofilm carrier for COD and BOD₅ reduction. It is believed that the novel EW system holds great promise as a low-cost wastewater-treatment system of choice, particularly in cases such as isolated or scattered settlements, agricultural and industrial effluents, private dwellings, hotels, parks and rural areas. At the same time, it offers a novel reuse alternative for the Al-WTS as opposed to landfill. Therefore, such a two-pronged approach towards alum sludge reuse would be environmentally and economically beneficial.

Setup of the Field Pilot-Scale EWs Trial

After extensive laboratory studies, a pilot-scale demonstration of an Al-WTS-based EWs system is currently carried out in an animal farm in Newcastle, County Dublin, Ireland, for the further development of such the novel EW. Engineering aspects, such as dealing with large amount of...
dewatered Al-WTS cakes in practice, clogging of EW systems, seasonal variation of temperature and open environment on the effect of the treatment efficiencies are under investigation. The setup of the field trial is shown in Fig. 3. The system consists of four identical plastic-wall wetland stages operated in series, with a total treatment surface area of 3.42 m². Dewatered Al-WTS cakes collected in fresh from Ballymore water treatment plant (Co. Dublin, Ireland) were used as the main substrate and total substrate depth in each cell is around 0.75 m. Loadings up to 0.29 m³/m²d (hydraulic) and 150.8 g-BOD₅/m²d (organic) have been applied across the entire system. Integrated animal farm wastewater (after primary settlement) from a research farm (with over 2,000 livestock units of sheep, pigs, cattle and horses) outside Dublin, was pumped from the feed tank (see Fig. 3) to the system via the first stage. The system is operated under the tidal flow strategy.

Results from the Field Trial

The overall COD removal performance of the system is illustrated in Fig. 4. During the startup period (25/02/2009–02/06/2009), the influent COD ranged from 57 to 1,087 mg/l with an average of 463 mg/l, while the effluent COD ranged from 97 to 790 mg/l. During this period, the COD removal was low as biological activity is being developed. The second period began from 6/5/2009. The influent COD ranged from 310 to 1,578 mg/l, while the effluent COD averaged 270 mg/l. The COD removal performance was improved significantly with average removal efficiency of 67.7%. It seems that the effect of temperature on COD removal is very limited. The removal efficiency remained around 67% even when the temperature dropped below 6°C in December 2009 as shown in Fig. 4. The average BOD₅ removal efficiencies of 51.4 and 66.8% were recorded for the startup and the second period, respectively (data not shown).

Figure 5 illustrates the overall performance on total nitrogen (TN) removal. The TN elimination is largely dependent on the influent BOD₅/TN ratio i.e., the available carbon source, as demonstrated in Fig. 5. During the start up period, the influent BOD₅/TN ratio was very low with an average of 0.9. According to Henze et al. [8], for complete denitrification the influent BOD₅/TN ratio should be close to 5. Apparently, the influent BOD₅/TN ratio during the start up period was far lower than this value. Consequently, the average TN removal efficiency of only 27% was achieved during this period. From 5 June to 17 August, the influent BOD₅/TN ratio increased significantly with an average of 2.41, due to the nature of the influent wastewater. As a result, the TN removal increased significantly from 50% to over 90% with an average of 64% removal over this period. However, from 21 August to 22 October the influent BOD₅/COD again decreased and this affected the TN removal, which dropped to 13.5–47.8%. All these results indicate that high TN elimination can be achieved within the novel EW, provided the carbon source is sufficient.

Performance of the EW system demonstrates a high and stable P removal, as illustrated in Fig. 6. Removal efficiency of above 90% was achieved within one week of operation. Thereafter, the removal efficiency was maintained in the range 82–100% under variable P loading rate caused by the nature of the influent wastewater. Removal efficiencies remained above 90% in most of the time with the average being 94.6% up to 12 November. The main pathway of P elimination is believed to be adsorption on the main substrate i.e. alum sludge. However, from 12 November the removal efficiency started to decrease and dropped to 73% on 10 December 2009. The reason may be that the alum sludge was starting to get saturated, because the P adsorption on the alum sludge has been going on for nearly 1 year under high P loading rate. This assumption can be confirmed during further operation of the CW.

Figure 7 illustrates the overall SS removal performance. During start-up period, the influent SS ranged between 27

| pH  | Q₀ (mg-P/g sludge) | Testing conditions | Reference                  |
|-----|--------------------|--------------------|----------------------------|
| 4.3 | 22.4               | P source: KH₂PO₄  | Yang et al. [14]            |
| 6.0 | 18.3               | Initial P concentration: 102 mg/l |                         |
| 7.0 | 14.3               | Equilibrium time: 48 h |                           |
| 8.5 | 1.1                | P source: (NaPO₃)₁₂–₁₃Na₂O Babatunde et al. [4] |
| 9.0 | 0.9                | Initial P concentration: 5.4 mg/l |                         |
| 4.0 | 4.5                | Equilibrium time: 48 h |                           |
| 6.0 | 4.1                |                   |                           |
| 7.0 | 3.1                |                   |                           |
| 8.0 | 2.0                |                   |                           |
| 9.0 | 1.7                |                   |                           |
and 485 mg/l with an average of 175 mg/l and the effluent SS was between 11 and 181 mg/l with an average of 77 mg/l. Accordingly, the removal efficiency varied between 17 and 83% with an average of 56%. In the second operation period, the influent and effluent SS ranged, respectively, between 31 and 633 mg/l with an average of 195 mg/l and between 0 and 221 mg/l with an average of 62 mg/l.

**Table 2 Summarized results of laboratory-scale model EW trials**

| Trial Conditions and Main Findings | Reference |
|-----------------------------------|-----------|
| Optimal configuration | The proportion of the Al-WTS in the four columns was 100, 80, 60 and 40%, respectively. The real animal farm wastewater was equally pumped to each of the column for 25 weeks over five P loading periods. The column with 100% Al-WTS showed the best for P removal and the same level for organics and SS removal compared with other columns tested. | Babatunde et al. [2] |
| Single EW trial | The single model EW is 100 cm in height and 14.5 cm in diameter. Common reeds, *Phragmites australis*, were planted on the top. It was operated in 'tidal flow' strategy for over 2 years with hydraulic loading rate of 0.5 m³/m²d and a range of organic loading rate of 11.5–143.5 g-BOD₅/m²d. The average removal efficiencies of 73.3 ± 15.9% for COD, 82.9 ± 12.3% for BOD₅, 86.4 ± 6.0% for RP (reactive P), 88.6 ± 7.2% for SRP (soluble reactive P) and 77.6 ± 17.5% for SS were achieved. The lifetime of the testing EW regarding the saturation of the Al-WTS for P immobilization was estimated as 2.5–3.7 years. | Zhao et al. [16] |
| Multi-stage EW system trial | The system consists of four identical single EW. Each was 90 cm in height and 9.5 cm in diameter and operated in 'tidal flow' strategy. Common reeds, *Phragmites australis*, were planted on the top of each EW. Real animal farm wastewater was fed to the system through the 1st to the 4th stage. Under a hydraulic loading rate of 1.27 m³/m²d and a range of organic loading rate of 279.4–774.7 g-BOD₅/m²d and 361.1–1028.7 g-COD/m²d, average removal efficiencies of 90.6 ± 7.5% for BOD₅ and 71.8 ± 10.2% for COD were achieved. P removal was exceptional with average removal efficiency of 97.6 ± 1.9% achieved for soluble reactive P at a mean influent concentration of 21.0 ± 2.9 mg/l. | Babatunde et al. [5] |
Discussion

The sustainable waste management into the future would embrace the concept of integrated waste management where the new challenge requires a very different response. The attempt of reusing dewatered Al-WTS in EW leads to the development of the novel Al-WTS-based EW, which is able to treat high P-containing wastewater. The dewatered Al-WTS used herein can be seen to have a significant and comparable P adsorption capacity (Table 1) compared with that of minerals, rocks, soils, marine sediments, industrial by-products and man-made products in which a P-removal capacity to vary from 0.025 to 32 mg-P/g was reported [11]. Pilot-scale trial in a farm provided convincing data to demonstrate that the dewatered Al-WTS is a promising material and therefore can be reused as a substrate in EW for wastewater treatment. Removal of COD and BOD$_5$ can be achieved at about the same level as in soil and gravel-based EW system [10]). This is also a good demonstration of the dewatered Al-WTS being a carrier for biofilm development in the EW and a suitable growth medium for planted reeds (Table 2; Fig. 3). More importantly, P removal can be significantly enhanced, and this provides evidence to show that the dewatered Al-WTS as a low cost adsorbent can considerably improve the P-immobilization capacity in EW system. Due to the fact that the Al-WTS is currently treated as a waste for landfill, the reuse of such sludge in EW will be reasonably recognised as a cost-effective solution in EW development with two-prong feature.

Available data from the first pilot-scale field system of Al-WTS-based EW has indicated the good tendency that the system is successful under the real situation.
More significantly, the system is a unique and promising low-cost wastewater treatment. Although technical parameters have been obtained from the field trial, which are useful for the purpose of design and operation, longer term operation is still desirable to validate the stability of the system. In particular, clogging of the system regarding the long term operation should be tested and the saturation of the alum sludge with P might also be usefully investigated over a more extended period before the large-scale application of the system.

**Conclusions**

This article describes the background of developing the Al-WTS-based engineered treatment wetland for enhanced treatment behaviour. Laboratory and pilot-scale field studies have all demonstrated the promise of such novel application of the ‘waste’ Al-WTS as a useful raw material employed in engineered wetland system for water pollution control. Batch P-adsorption tests revealed that the Al-WTS tested possess excellent P-adsorption ability, which forms the basis of further investigation. Trials on treatment wetland with Al-WTS as main substrate indicate that the dewatered Al-WTS can be a carrier for biofilm development and a good medium for wetland plant growth. The development so far supports the proposition that the potential reuse of dewatered Al-WTS as a substrate in engineered treatment wetland system can be a promising solution to transfer Al-WTS as a ‘waste’ to a useful raw material, in developing a cost-effective treatment wetland system. Such the development would be environmentally
and economically beneficial, with obvious feature of two-prong characteristics.

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