



Valorization of undervalued aluminum-based waterworks sludge waste for the science of “The 5 Rs’ criteria”

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Abstract

Alum sludge (AS) is an underrated by-product waste resulted from treating raw water through conventional water treatment plants. Water is attained from various reservoirs such as rivers and aquifers, and it may contain a wide variety of contaminants as drinking water processing systems produce “waste” sludge as a residual material that causes significant environmental issues. Hence, numerous efforts aimed to sustainable reuses of such sludge. This article illustrates the beneficial reuses of the aluminum-based sludge to close the loop between the sludge waste generation and the sustainable environment with providing past, current and updated knowledge with the introduced uses with their advantages and challenges. Alum sludge as a resource not as a “waste” for reuse facilities is considered a value-added alternative for management achieving the legislation requirements and proposing “end-of-waste” concept. 5Rs, including “Reduce, Reprocess, Reuse, Recycle and Recover,” are providing sustainable solution to fulfill present and future aspects for green environment and safe sludge disposal. In this regard, now, it is essential to focus and adopt the 5Rs criteria for the concept of circular economy that replaces the “end-of-life” principal with restoration.

Keywords Alum sludge · Conditioning and dewatering · Reprocess · Reuse · Wastewater treatment · Constructed wetland

Introduction

The undervalued materials that arise as a result of numerous human activities (i.e., domestic agricultural, industrial etc.) are unwanted solid waste materials. They may cause a direct deterioration to the environment, and they could be a reason for a land pollution as well as water contamination (Tony and Ali 2021). Therefore, in the modern societies, for managing such unwanted solid wastes, the 5Rs “Reduce, Reprocess, Reuse, Recycle and Recover” principle is introduced. Such criteria are applied to utilize the waste materials into support sustainable environment, minimize various environmental pollution types, diminish the consumption of unnecessary substances and promote the maximal lifetime utilization of the products. Reduce includes minimizing the unnecessary use solid waste. To add up, reduce could be

attained by reclaiming the waste by repressing and reusing it for different purposes. Thus, it increases the lifetime of waste and reduces its production. Hence, this preserves the environmental resources. However, recovering the previously used waste materials and introducing a new life from it is termed as recycle. Processing and modifying the waste into another form is so-called recycling the waste. This could be applied in such a case that the waste could not possess the potential of reuse and reduce. Recover is characterized as processing the waste in a way that saves it from being destroyed. With the aim to achieve economical savings of waste disposal for materials and maintain the environment sustainably option, 5 Rs’ criterion is a suitable alternative for waste management (Zhao et al. 2021; Zhu et al. 2021). Thus, alum sludge as a solid waste is introduced to the 5Rs criteria.

Worldwide, potable water is a prerequisite as well as its quality is cardinal as a result of urbanization and booming population. Therefore, a water-related technology for the production of reliable and safe drinking water treatment plants is a must (Ashour et al. 2014; Thabet et al. 2020; Zhang et al. 2021). Therefore, utmost priority has been attained globally to receive a clean drinking water. In this regard, integrated numerous methodologies have

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been applied in the typical drinking water treatment plants (Thomas et al. 2016; Yu et al. 2021). Thus, different unit operations are used, in which coagulation and flocculation operations chemicals are used as primary coagulants or as coagulant aids. The result is a proportional amount of by-product residual sludge is generated that is essentially required to be subjected through a dewatering process before its final disposal in order to its thickening (Gregory and Duan 2001; Carty et al. 2002; Ashour and Tony 2017).

In some countries, such as Egypt, the disposal technique of alum sludge is the direct disposal into nearby drains that meets the water sources intake (Tony and Tayeb 2016). In other places, it could be discharged into open lands areas which is considered a simple and cost-efficient of sludge disposal technology (Vasudevan; Thabet et al. 2021). However, this is not considered ideal solutions since such techniques cause contamination of water streams and soil from the chemicals disposed off with the sludge. Hence, efficient alum sludge management in an eco-friendly way remains an issue for researchers. The answer is the 5 Rs' "Reduce, Reprocess, Reuse, Recycle and Recover" of the sludge which could offer a sustainable management solution of such waste.

In the available literature, abundant research is cited dealing with sewage sludge utilization (Fytily and Zabaniotou 2008; Kelessidis and Stasinakis 2012) although the review studies regarding waterworks sludge utilization and reuse and applications are not many and recent (Babatunde and Zhao 2007; Dassanayake et al. 2015). Therefore, in order to expand such previous investigations and to generate a comprehensive outlines of the alum sludge production and possible options for end possibilities of reuse, the main target behind this work is to present a comprehensive review of sustainable approaches and prospective trends in waterworks sludge handling and management for its beneficial reuses. So, this study also deals with highlighting the particular focus on numerous studies that are dealing with the alum sludge applications such as land applications (Dassanayake et al. 2015), construction materials enhancement and wastewater treatment applications. The barriers stands over the widespread of such reuses will be also explored. Understanding and identification of the main gaps and issues surrounding the potential environmental impacts are included. Likewise, the bibliometric data analysis were performed through this investigation, via VOSviewer software, the key terms, title and abstract were incorporated to the software in order to create a term co-occurrence mapping and density visualization.

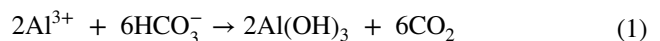
In the line of the 5Rs' principle for sustainable development, alum sludge has been intensively studied in the last

decades as a value-added material. On that basis, converting undervalued alum sludge waste into useful product improved alum sludge into more expansive and improved alum sludge reuse routes.

Alum sludge production and quantities

Incorporated coagulation along with filtration (Konieczny et al. 2009; Yin 2010) and sedimentation (De Sena et al. 2009; Amaral Filho et al. 2016) where the water quality is a superiority (WHO 2011) is applied in the drinking water treatment facilities. Chemical purification methodology that is signified as coagulation process is categorized to be an expensive technology since the costive chemicals applied (Teh et al. 2016; Yu et al. 2017; Sillanpää et al. 2018; Tayeb et al. 2019). In such technique, coagulant aids that is usually depends on metal-based coagulants, i.e., Al and Fe, are used to precipitate the soluble metal ions in water (schematic representation of the drinking water treatment steps is displayed in Fig. 1). But, special concerns have been gained scientists attention since massive amounts of sludge including heavy metals residuals are generated from such processes. Such sludges are categorized as potentially toxic to the aquatic environment (Tetteh and Rathilal 2019; Thabet et al., 2021a).

On the other hand, a technical generation of a proportional amount of by-product sludge is produced which is brought through a dewatering process. It is passed through treatment steps before the final disposal to reduce its amounts (Zhao et al. 2018; Tony and Lin 2020a). Such generated by-product is aluminum-rich-based sludge which is so-called alum sludge. Alum sludge is generated as a result of adding aluminum sulfate [$Al_2(SO_4)_3 \cdot 14H_2O$] as a flocculating agent in the drinking water treatment process (Eq. 1) (Ahmad et al. 2016). The resultant alum sludge is a two-phase mixture of solids and water, and its water content is generally in the level between 99% (before thickening) and 95% (after thickening). Such sludges are often referred as 'difficult-to-dewater' (Goldman and Watson 1975; Zhao and Bache 2001; Simpson et al. 2002; Zhao et al. 2018).



Virtually, alum sludge that is produced from drinking water treatment plants signifies up to 3% the volume of the raw water that is subjected for treatment (Hidalgo et al. 2017). However, it is difficulty to attain an exact estimation to the alum sludge produced in a region or a specific country. Also, for the European union (EU) countries, the alum sludge is categorized as Code 19's list of waste, so it

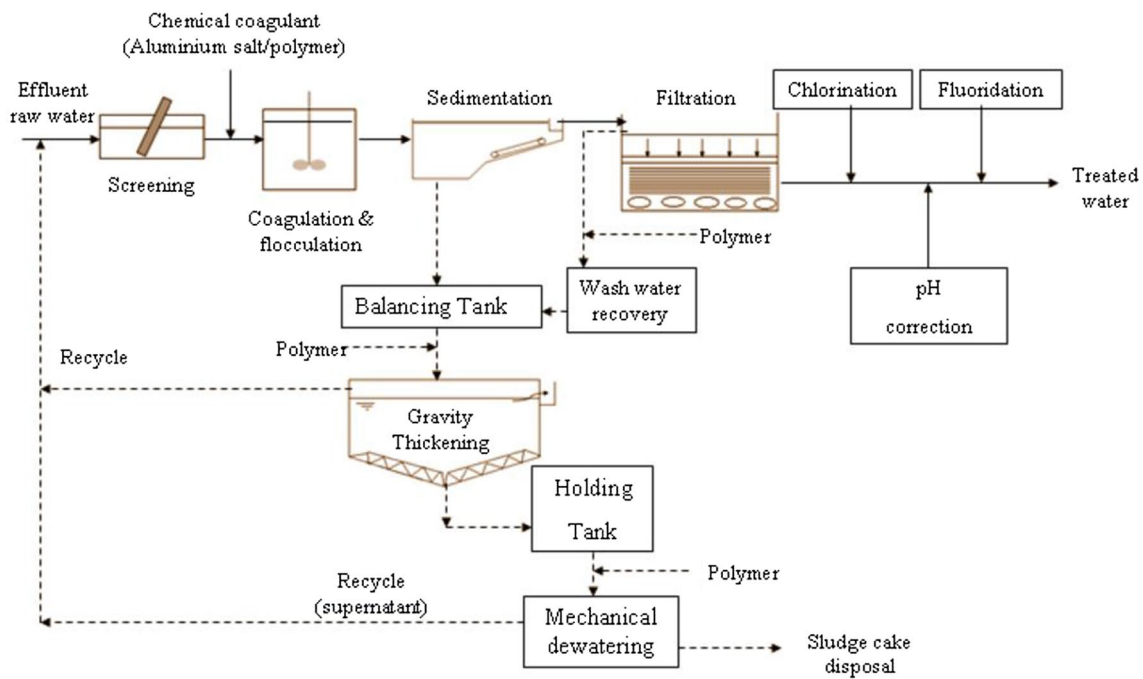


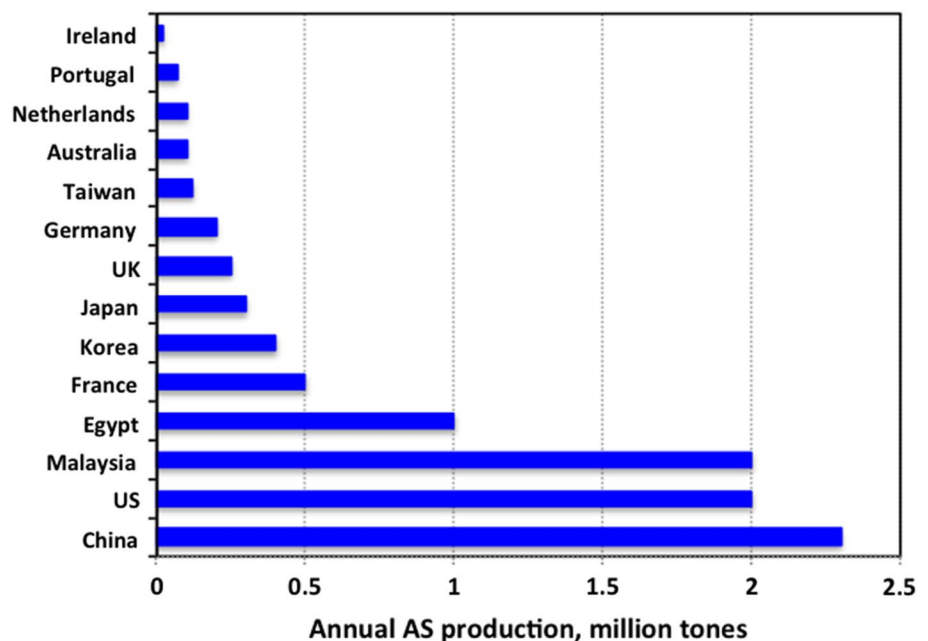
Fig. 1 Schematic representation flowchart of watercourses treatment plant and sludge stream in water works plant (adapted from: EPA, 2002; Gregory and Dillon, 2002; Mazari et al. 2018; Zhao et al. 2021)

is difficult to attain a published data about the waste statistics according to the local authorities (Renou-Wilson et al. 2019). Therefore, it is unsurprisingly that it could not attain a specific on regional and consequently the global alum sludge waste data.

Annually, the generated alum sludge in numerous countries is estimated and illustrated in Fig. 2, which summarizes

the data available in the two decades (Foss and O’Connell 1996; Babatunde et al. 2007; Xiong and Mahmood 2010; Hu et al. 2012; Dassanayake et al. 2015; Ahmad et al. 2016; Abd El-Razek et al. 2019).

Fig. 2 Estimated alum sludge production quantities in some selected countries



Bibliometric analysis

Recently, bibliometric analyses were emerged as a useful tool for analyzing and identifying a certain subject and mapping key aspects. The most cited items related to alum sludge and their related terms are investigated via bibliometric mapping. Therefore, to attain a state of the art outline of the alum sludge, the literature survey was performed on the Web of Science platform. The search term was “Alum sludge OR Aluminium sludge OR Waterworks sludge”. Then, the information revealed from “Web of Science Core Collection” database was extracted through the period of 2000 to August 2021.

The relatively limited articles in the last two decades attained from the Google Scholar search platform are given in Fig. 3 to illustrate a profile of the current statuses of the studies conducted and published related alum sludge. Overall, articles were extracted and displayed as annually cumulative publication numbers jointly with the alum sludge application profiles. The number of research studies according to Fig. 3 is in increase which gives the opportunity of the research and development (R&D) of the alum sludge to its maximal level of reuse facilities to reach to a satisfied reasonable sustainable disposal. A brief overview of the studied research investigations represents popularity of this technology application as referred in the sub-plotting in Fig. 3. A wide range of application include wastewater treatments with varies reuse purpose comprising adsorption (Liu et al. 2021a, b), coagulation and wastewater treatment facilities. Also, alum sludge could be used as a building material in

cement industry enhancement or brick making. Moreover, numerous studies as well dealt with the land applications such as the agriculture uses and as a substrate in constructed wetland (CW).

Web of Science platform and the VOSviewer software (version 1.6.16.0, accessed on 15 August 2021) were applied for this design purpose and to analyze the keywords of the papers. In brief, the designed attained maps are incorporating network, and overlay and density visualization mapping are designed through the software according to the general procedure incorporating: (1) downloading the search data attained from “Web of Science” platform; (2) the attained data from Web of Science are saved in the required format and inserted to VOSviewer software; (3) analysis is conducting through the required method. In the current article, the co-occurrence associated with authors’ keywords was selected to be the minimum number of occurrences. Based on the program manual, mapping was subsequently designed after adjusting the parameters in the software. Overlay visualization is one of the most common features of the VOSviewer to display density visualization over time periods. Alum sludge research is conducted that extracted from the cited research articles, and the bibliometric mapping of clusters can be identified as seen of Fig. 4. The hot-spots clusters could illustrate the intensive research studies based on 1509 results obtained through data from “Web of Science” core collection from the search terms “TI is (alum sludge OR aluminium sludge OR waterworks sludge)”.

To add up, extra data analysis was conducted via software (Fig. 5) displaying overlay visualization of the bibliometric mapping. The big dot areas signify hotspots of analysis. The

Fig. 3 Joint plotting of number of publications during the last two decades regarding alum sludge and the cumulative publication of the type of alum sludge applications (data based on Google Scholar Search on August 2021)

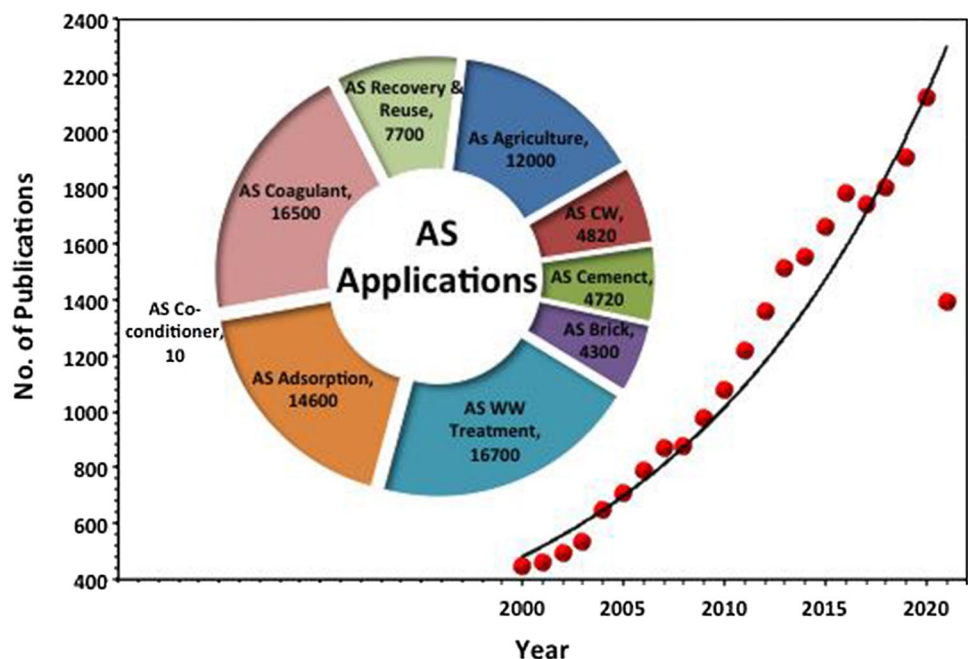


Fig. 4 Keyword density visualization of alum sludge research work in the last two decades based on “Web of Science” platform to signify the research clusters using 1509 results obtained through data from “Web of Science” core collection from the search terms “TI (alum sludge OR aluminium sludge OR waterworks sludge)”

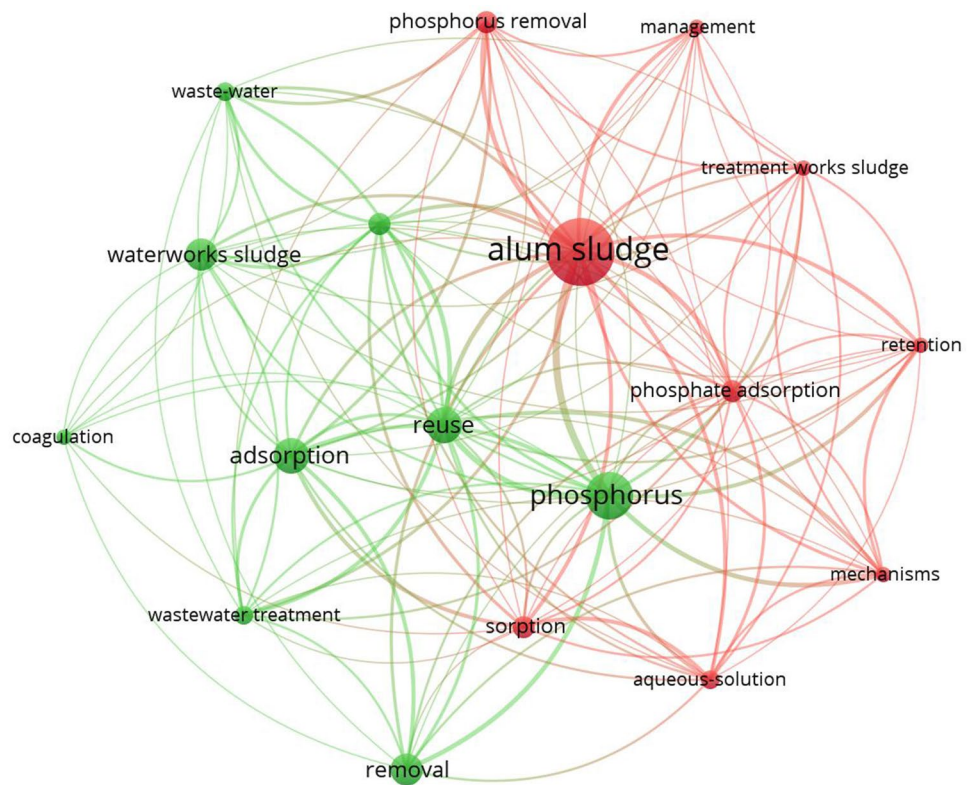
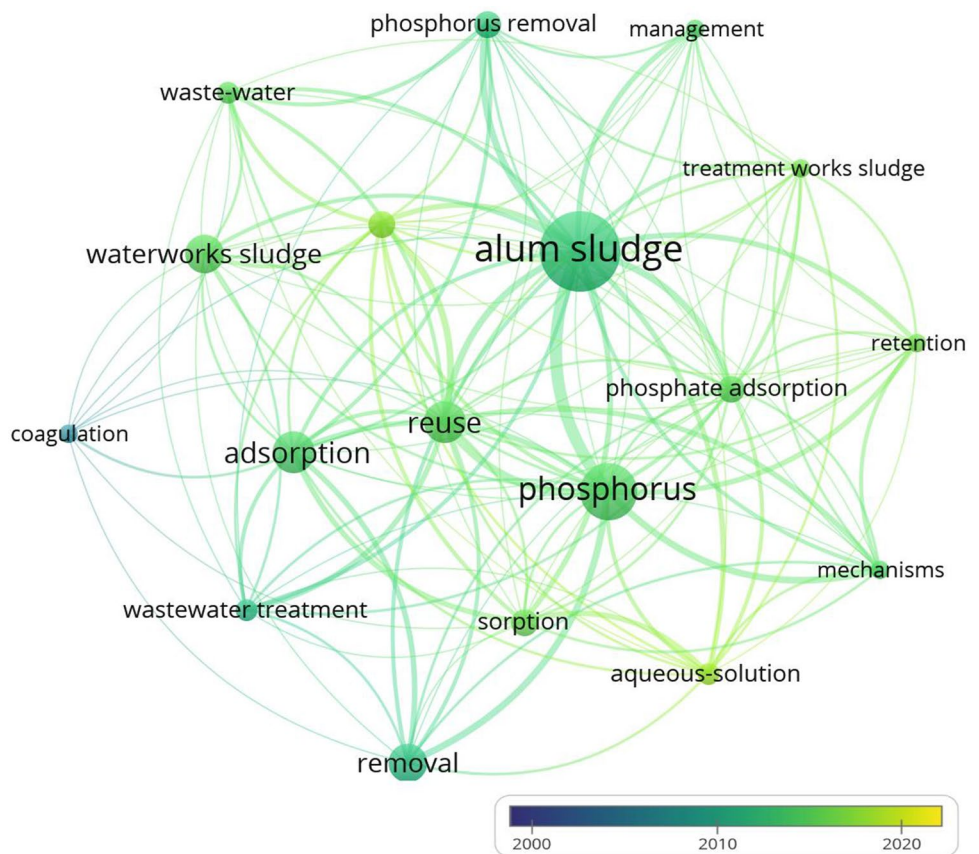


Fig. 5 Bibliometric research mapping through the last two decades based on overlay density visualization



size of clusters is related to the research importance and significance in the area. As seen in the legend in Fig. 5, in the 2000s, focus was placed on adsorption studies of phosphorus and other elements, while in the 2010s, adsorption mechanisms and the development of reuse strategies were the main focus. In the 2020s, reuse in wastewater treatment technology became more accepted in AS studies.

Composition and physicochemical characterization

Recent lifestyles are requiring more water consumption per capita, so fresh water demand is in increase. Further, the urgent need for clean water is a prerequisite. In such context, drinking water treatment plants produce massive amounts of waste by-product, which is so-called waterworks sludge. Aluminum sulfate is usually used as a primary coagulant in drinking water treatment plants, the result is that aluminum ions were hydrolyzed into aluminum hydroxide that is precipitated, and the so-called alum sludge (AS) is generated (Basri et al. 2019). Insoluble water impurities and organic substances are absorbed by aluminum hydroxide through raw water processing. Typically, the solid content of the thickened sludge is about 2–4%; however, the solids could be reached to 17–23% after mechanical dewatering through centrifugation.

Alum sludge feature is subjected to the raw water quality and the chemicals grades used specially the alum purity. Consequently, the physicochemical characteristics and geotechnical properties of the AS may be widely varied. Based on the available literature cited, the physicochemical characteristics and geotechnical data range of the alum sludge are displayed in Table 1. Such data reveal a wide range in most characteristics this is mainly due to the different sources of alum sludge waste, which is based on the source of watercourse water (Babatunde et al. 2007; Ippolito et al. 2011; Ren et al. 2020). Moreover, alum sludge properties are highly characterized by the variation in the coagulant applied in the water treatment plant. The solid content in the alum sludge varies from one water treatment plant to another, as the type of the raw water treated is varies in the solid concentration and the turbidity. The solid content of the sludge from ordinary different waterworks steps are varied. The sludge outlines the sludge blanket clarifier containing 0.1 to 0.5% solid content. However, in the gravity thickening tank (within 3 to 4 months) contains 3–4% solid content. This could be increased to 4 to 6% when polymer is added. Also, if the sludge dried, the solid reached 45% (Ahmad et al. 2016; Rebosura Jr et al. 2020; Zhao et al. 2021). In contrary, the resultant sludge from the sedimentation/coagulation process has a low solids concentration which makes it difficult to dewater (Tony 2020b).

Assessment of alum sludge toxicity

According to the available literature cited, there is insufficient information regarding the potential toxicity of drinking water plants residual sludge. Also, the available information is limited and supplies some conflict. Hence, critical research studies assess alum sludge toxicity from several decades. Aluminum is very toxic, both acute and chronic terms, to aquatic habitat in water, especially in low pH levels. This paper provides a description of ecological risk assessment that is based on the framework recommended by United States Environmental Protection Agency (USEPA), which includes problem formulation, characterization of exposure and its effects and risk characterization.

George et al. (1995) stated the alum sludges discharged from the several waterworks plants in North America into the waters might affect algal growth. In contrary, Skene et al. (1995) revealed that the Al-toxic effect is not evident since alum sludges were applied as growth media. Likewise, other study by Sotero-Santos et al. (2005) did not report an exact toxicity evidence of alum sludge on short-term exposure; however, low toxicity is attained through the long-term contact, while the wide range of sludge characteristics is not related to the degree of toxicity. Further research was conducted by Dayton and Basta (2001) concluded that heavy metal levels in the waste alum sludge are less than the regulatory limits. Similar data are highlighted by Elliot and Dempsey (1991). However, Förstner and Haase (1998) investigation reported that the metals from alum sludge are a pH-dependent process. According to the ecological risk assessment provided by United States Environmental Protection Agency (USEPA), aluminum is very toxic to the aquatic habitats in ecological system (Mortula et al. 2009). Wang and He 2010 reported that aluminum concentration in the Zaohe River in USA and Canada and could do harm to human health.

Hence, lack of reasonable qualitative and quantitative assessment available and the magnitude of the potential toxicity effect of alum sludge is alarming to do more research to investigate a rational data.

Evaluating the sludge dewaterability

CST: Capillary suction time, CST, and specific resistance for filtration, SRF, are the most known methods to evaluate the sludge dewaterability as quantitative indexes for the evaluation of the dewatering performance. CST test has been used since the 1970's as a reliable, inexpensive, easy and a rapid method for characterizing filterability and dewaterability of a given sludge; hence, it is considered a pragmatic index. CST stands for the time required for sludge to complete its

Table 1 Physicochemical characteristics and geotechnical properties of alum sludge*

Parameter	Unit	Range	References
pH	–	5.12–8.4	Ippolito et al. (2011), Oliver et al. (2011), Putra and Tanaka (2011), Ulén et al. (2012), (Castaldi et al. 2014) Castaldi et al. (2014)(Rebosura Jr et al. 2020) (Razali et al. 2007) Rebosura et al. 2020; Ahmad et al. 2016; Tony et al. 2008; Yang et al. 2006; Razali et al. 2007
TSS	mg/L	1450–52,354	Rebosura et al. 2020; Ahmad et al. 2016; Yang et al. 2006; Razali et al. 2007
VSS	% of TS	29	Nabarlatz et al. 2010; (Anbazhagan and Palani 2018)
COD	mg-COD/L	216–226	Dassanayake et al. 2015; Tony et al. 2008
BOD ₅	mg-BOD/L	45–104	Castaldi et al. 2014; Rebosura et al. 2020; Rebosura et al. 2020
CST	s	67.5	Tony et al. 2008
SRF	m/kg	19.8×10^{10} – 2.24×10^{13}	Tony et al. 2008; Sun et al. 2015; Tony et al. 2016
DS	%	3	Tony et al. 2016
ζ P	(mV)	– 22.0 – – 10.8	Pan et al. 2003; Mazari et al. 2018; Tony 2020a, b
Conductivity	Ms/cm	600 -650	Mazari et al. 2018
SG	–	0.95–2.34	Foroughi et al. 2018; Breesem et al. 2014
<i>Chemical analysis</i>			
Aluminum	g-Al/kg	7–142	Rebosura et al. 2020; Agyin-Birikorang and O'Connor (2009), Hovsepyan and Bonzongo (2009), Makris et al. (2006), Ippolito et al. (2011); Babatunde and Zhao (2007); Ippolito et al. (2011), Ren et al. (2020)
Iron	g-Fe /kg	4.87–37	Ippolito et al. 2011, Razali et al. 2007, Rebosura et al. 2020
Calcium	g-Ca /kg	1.825	Ren et al. 2020
Magnesium	g-Mg /kg	1.751–2.979	Babatunde and Zhao 2007, Ippolito et al. 2011, Rebosura et al. 2020, Ren et al. 2020
Manganese	g-Mn /kg	1.977	Ren et al. 2020
Sodium	g-Na /kg	0.212–1.541	Chiang et al. 2009, Rebosura et al. 2020, Ren et al. 2020
Potassium	g-K /kg	1.751 -7.149	Sales et al. 2011, Rebosura et al. 2020
Sulfur	g-S /g	2.24 -9.72	Rebosura et al. 2020 Rebosura et al. 2020, Ren et al. 2020
Silicon	g-Si /kg	134.041	Ren et al. 2020
Cobalt	g-Co /kg	0.001	Sales et al. 2011, Rebosura et al. 2020
Phosphorus	g-P /kg	0.059–0.062	Castaldi et al. (2014); Rebosura et al. 2020; Ahmad et al. 2016; Yang et al. 2006; Razali et al. 2007; Rebosura et al. 2020; Rebosura et al. 2020, Ren et al. 2020
Nickel	g-Ni /kg	0.061–0.082	Rebosura et al. 2020 Rebosura et al. 2020, Ren et al. 2020 Razali et al. 2007 Rebosura et al. 2020 Rebosura et al. 2020
Copper	g-Cu /kg	0.001–0.07	Rebosura et al. 2020 Rebosura et al. 2020, Ren et al. 2020 Razali et al. 2007; Rebosura et al. 2020
Lead	g-Pb /kg	0.0059–0.082	Rebosura et al. 2020 Rebosura et al. 2020, Ren et al. 2020 Rebosura et al. 2020
Chromium	g-Cr /kg	0.015–0.045	Rebosura et al. 2020 Rebosura et al. 2020, Ren et al. 2020 Rebosura et al. 2020

*TSS Total Suspended Solids; VSS Volatile Suspended Solids; SG Specific Gravity; ζ P Zeta potential; SRF specific resistance to filtration; CST capillary suction time; DS dry solid

filtration (Yin et al. 2004; Scholz 2005). Although the CST is a good index for the solid concentration of the sludge, it cannot estimate directly the bound water for the sludge (Yin et al. 2004). However, CST can be used as an index for evaluating of the average SRF of the evaluated sludge (Ma et al. 2007). Investigators results reported that alum sludge from the water works is slightly more difficult to dewater than some wastewater sludge since waterworks sludge ranged CST value ranged from 57.3 to 84 s (Zhao and Bache 2001;

Zhao 2004); however, biological sludge showed about 30 s for CST (Mikkelsen 2001).

SRF: Also, specific resistance for filtration, SRF, is a sludge dewaterability index through evaluating the extent of water yield via filtration procedure. SRF is a useful method to evaluate the dewaterability of the sludge and to optimize the coagulant dose. SRF is established on the relation between sludge viscosity and the decrease of pressure over a certain distance. The standard SRF test was performed using simple lab test technique. Generally, the sludge type

governs the SRF values since numerous studies recorded various sludge values according to the sludge type used. For instance, Zhao and Bache, (2001) represented that the raw alum sludge from the drinking water treatment plant ranges from 63.2 to 133.4×10^{12} m/kg. Ma et al., (2007) recorded it as 7.3×10^{12} m/kg. Lu et al. 2001 reported that the original activated sludge is 1.17×10^{13} m/kg, while (Buyukkamaci 2004) Buyukkamaci, (2004) reported 9.162×10^{10} m/kg is the SRF value of the sludge used through such study.

Christensen et al. (1993) correlated a relation between CST and SRF (Eq. 2):

$$\text{CST} = \text{SRF} \times c_1 \times \mu + w \times c_2 \times \mu \quad (2)$$

where c_1 and c_2 are coefficients related to CST, μ is the viscosity of the filtrate, and w is the solid content in unit volume of the filtrate.

Although CST and SRF are the most widely reliable used techniques to evaluate sludge dewaterability, there are other methods available for measuring sludge dewaterability.

DS: dry solids (DS) content (Pan et al. 2004; Dewil et al. 2005): DS technology evaluates and measures the DS content in the sludge cake after evaporation (at 105 °C).

Bound Water: Generally, the aqueous phase in the sludge is classified into two main categories, free water and bound water, whereas according to (Yin et al. 2004), the water in sludge is classified according to their bonding to the solids in the sludge. The classification falls into free water and bound water (Yin et al. 2004). Free water is not attached to sludge solids and could be separated by simple gravitational settling or mechanical dewatering. However, bound water can be broken down into: (i) interstitial water, water that is trapped inside the floc structure by capillary forces or within a cell; (ii) surface water, water held onto the surface of solid particles by adsorption and adhesive forces; and (iii) chemically bound water (intracellular water): water that is chemically bound to the particles and it cannot be released easily by mechanical means; and it can be released only by applying heat to help in the thermal chemical destruction of the particles. Centrifugation method, dilatometric measurement and differential scanning calorimetry could measure the bound water concentration in sludge (Yin et al. 2004).

Zeta Potential: this is a physical measure technique of the charge on a colloidal particle and presents the surface characters of sludge (Yin et al. 2004). Such measurement is dependent on the repulsive force between sludge particles and the distance over the particles that may repel each other and thus prevent coagulation. The high positive or negative values of zeta potential mean that the particles are stable and hard to coagulate. In contrary, the low positive or negative zeta potential means that the particles are unstable and easy to coagulate (Yin et al. 2004). – 17.44 to – 37.4 mV is

reported to be the possible zeta potential of the alum sludge (Guan et al. 2005).

Sludge conditioning/dewatering for disposal practices

The whole sludge management trained is commonly comprising of a series of generally divided into successive five treatment steps. Such successive steps, namely thickening, stabilization, conditioning/dewatering and the final disposal or reuse facilities. Among them, water is removed from sludge solids through thickening and dewatering procedures in which the volume of water is deduced and hence the sludge volume is reduced to be ready for treatment in the further processes. Conditioning is subjected for the object of improving the sludge dewaterability. Conditioning could be achieved through chemical aids such as flocculants, acid, ferric chloride and lime addition or through physical disruption. Finally, before the sludge disposal it is subjected to mechanical dewatering in which press filter, centrifuges or dryers are applied to reduce the water content in the final sludge to about 80% including a range of (20–25%) of dry solids, DS in the dewatered sludge cake (Yin et al. 2004).

Conditioning and dewatering techniques gained scientists and researchers great attention due to the challenges of the massive sludge produced. Extensive research has been conducted that are dealing with sludge conditioning and dewatering to satisfy the more stringent rules and restrictions appeared by the authorities and legitimations. Thus, reasonable conditioning and dewatering performances to attain higher sludge disposal and reuse facilities in order to reach to a minimal environmental impacts are ever-increasing (Tony et al. 2008; Zhou et al. 2014).

Numerous methodologies incorporating both physical, i.e., heat, freezing and mechanical treatments, and/or chemical treatment amenities are extensively applied to such sludge handling and conditioning to improve its dewaterability. However, chemical treatments still more pronounced effect and involve energy-saving advantages which include adding the flocculating agents, acids and alkaline (Zhou et al. 2014). In the recent decades, advanced oxidization conditioning technologies comprising Fenton's reagent have been applied as a non-polymer conditioning techniques. Not only energy-saving of the chemical conditioning in comparison to the physical conditioning its merits, but also, the oxidization processes are potentially removing recalcitrant compounds in such sludge. Thus, the final sludge disposal to the environment is more environmentally benign (Ma et al. 2007).

Chemical conditioners that are based on advanced oxidation processes are gaining a great scientist' attention. Fenton's oxidation is a promising superior dewatering technique

possessing numerous advantages (Tony et al. 2008). Although the Fenton oxidation is a superior conditioning process compared to the other chemical conditioners, a few research cited are dealing with such investigation (Zhao et al. 2009). Therefore, there are still significant research gaps at present and more research is required to examine the relationship between the needed sludge oxidants and dewaterability and optimization of the operational parameters.

The data tabulated in Table 2 reveal that the scattered limited studies dealt with alum sludge collected from different regions of water treatment plants (WTP) as a by-product residual from waterworks. Sludge dewatering showed that polymer dewatering displayed higher treatment efficiencies than other conditioners. However, it is noteworthy to mention that, the toxicity related to polymers stands as a problem. Therefore, scientists are dealing with searching for alternatives. Generally, low toxicity is attained for the normally applied anionic and non-ionic polymers. But, cationic polymers types found to be more hazards to the eco-system and aquatic organisms. Hence, there are strict limits on their use in the drinking water treatment plants for preventing their existence over the permissible limits to stop the environmental damage. Notably, monomers used in the polymer production showed toxicity more than the polymer itself; nonetheless, severe quantities on their levels are sustained, especially with regard to acrylamide. In this regard of rigorous regulations on the polymer uses, polymer toxicity could not seem to be a big problem (Bolto and Gregory 2007).

Additionally, it is significant to mention that not only the environmental aspects are stands behind searching for polymer alternatives, but also the economic prospective. Even the most cost-efficient polymer types are not considered from the industrial potential of a lowest cost (Tony et al. 2008). Notably, it is recorded by Ghebremichael and Hultm (2004), 65% of dewatering is achieved through SRF and CST reduction recording via using *Moringa oleifera* (MO). It is a significant research which concluded that MO could be effectively used and replaced alum or polyelectrolytes for chemical conditioning of water works sludge to prepare it for dewatering. It is recommended as a green alternative option. Moreover, the study introduced by Pan et al. (2004) explored that minimal effect could be attained when surfactant is applied as conditioner in comparison to polymer; however, no effective distinguish is achieved with types of the surfactant charge.

Moreover, it is significant to illustrate that dual polymer conditioning does not demonstrate a considerable improvement and further reduction in the sludge dewaterability and filterability assessed via CST and SRF comparing it with the solo polymer conditioning as seen the results in Table 2 by Ma et al. (2007). Hence, considering dual conditioning achievement should notably consider both cost and

administration, which is not a recommended strategy for the real industrial scale applications.

As a further bonus, the oxidation conditioners (i.e., Fenton reaction) are less aggressive to the environment since their reactions are environmental benign than organic polymers, resulting in a green and sustainable ecosystem. Although alum sludge conditioning by advanced oxidation process specially Fenton's oxidation is not superior as polymer conditioning for improvement of sludge dewaterability, its environmentally benign compared to polymeric addition should be considered. Moreover, till now, most of the researches conducted are mainly focused on the application of classical Fenton peroxidation reaction according to the data displayed in Table 2. Additionally, only limited pilot-scale tests had been introduced. Hence, data are still lacking for large-scale applications results, in which optimization and pilot-scale tests are required. Also, there is limited research conducted on alternative oxidization processes such as different iron sources or other transition metals oxidization process and ozonation process. Furthermore, basic mechanisms of alternative advanced oxidation processes should be clarified and studied in more detailed. The influence of more effective or enhancing the catalytic oxidation conditioners performance still requires more study. Moreover, detailed research is needed dealing with the mechanism on their chemical performance that has as yet only been investigated superficially.

Sustainable reuse of waterworks sludge residues

Research into a sustainable disposal opportunity is the scientists and researchers job to reach a green ecosystem. Hence, alternative waterworks sludge disposal option to maximize their value and improve their benefits and reach these massive amounts from underrated material to a value added product is crucial research work to satisfy the restrictions and limitations. In this regard, alum sludge is practiced to numerous applications according to the following trails: (1) building material; (2) agriculture applications; (3) adsorbent material; (4) coagulant material; (5) co-conditioner and (6) constructed wetland applications. Figure 6 summarizes varies beneficial reusing and reprocessing applications of AS for sustainable endpoint solution.

Building materials

Soil stabilization before road infrastructure

Ever though the aforementioned difficulties associated with alum sludge handling as a waterworks residue, just limited research articles have tried to valorize it as a building and

Table 2 Summary of relevant studies on alum sludge conditioning via various chemical conditioners capabilities

AS, Country of study	Conditioner ^a	Dosage ^b	TSS, mg/L	pH	Tim. min ^b	CST reduction, %	SRF reduction, %	DS increase, %	Ref
Collected from North Richmond conventional WTP, Australia	Cationic polymer, Zetag-89	3 kg/t-DS	–	7.3	–	–	–	20.4	Dharmappa et al. 1997
Collected from Nepean Dam WTP, contact filtration plant, Australia	Anionic polymer, Magnafloc LT-25	6 kg/t-DS	–	7.3	–	–	–	6	Dharmappa et al. 1997
Collected from North Richmond conventional WTP, Australia	Cationic polymer, Zetag-89	3 kg/t-DS	–	7.3	–	–	–	9	Dharmappa et al. 1997
Collected from WTP, Nepean Dam (ND), Australia	Anionic polymer, Magnafloc LT-25	5 kg/t-DS	–	7.3	–	–	–	6	Dharmappa et al. 1997
Collected from the clarifiers at Burncrooks WTP West of Scotland, UK	Anionic polymer, Floccimiser 50	3.85 mg/L	1,500	6.3	–	70	85	–	Keenan et al. 1998
Collected from bottoms of settling basin & concentration tank, Feng-Yuan Taichun WTP, Taiwan	Cationic surfactant CTAB	2 mg/L	–	–	60	57	66	4%	Ruhsing Pan et al. 2000
Collected from bottoms of settling basin & concentration tank, Feng-Yuan Taichun WTP, Taiwan	Anionic surfactant SDS	1 mg/L	–	–	60	65	66	3%	Ruhsing Pan et al. 2000
Collected from sludge holding tank WTP, UK	Polymer, Magnafloc LT25	20 mg/L	9300 mg/L	6.6–7.3	1.5	–	–	30%	Zhao and Bache 2002
Collected from sedimentation tank, Feng-yuan WTP Taichung, Taiwan	Cationic polymer, PC-320	20 mg/L	–	–	1	–	85	–	Wu et al. 2003
Collected from concentration basin, Hsin-Chu WTP, Hsin-Chu, Taiwan	Cationic, Polymer, PC-325	–	60,000	7.05	1	96	58	98	Pan et al. 2003
Collected from concentration basin, Hsin-Chu WTP, Hsin-chu, Taiwan	Chitosan	–	60,000	7.05	1	88	21	98	Pan et al. 2003
Collected from sedimentation tanks, Lov'oo WTP, Stockholm	Cationic polyelectrolyte Praestol 650 TR	1.8 kg/t	33,900 mg/L	6.10–6.40	30	96	96	–	Ghebremichael and Hultman 2004
Collected from sedimentation tanks, Lov'oo WTP, Stockholm	Anionic polyelectrolyte, Praestol 2540 TR	1.8 kg/t	33,900 mg/L	6.10–6.40	30	93	92	–	Ghebremichael and Hultman 2004

Table 2 (continued)

AS, Country of study	Conditioner ^a	Dosage ^b	TSS, mg/L	pH	Tim, min ^b	CST reduc-tion, %	SRF reduc-tion, %	DS increase, %	Ref
Collected from sedimentation tanks, Lovö WTP, Stockholm	Alum	63 kg/t-DS	33,900 mg/L	6.10–6.40	30	76	81	–	Ghebre-michael and Hultman 2004
Collected from sedimentation tanks, Lovö WTP, Stockholm	MO	125 kg/t-DS	33,900 mg/L	6.10–6.40	30	66	65	–	Ghebre-michael and Hultman 2004
Collected from sludge holding tank Ballymore Eustace WTP, Southwest Dublin, Ireland	Gypsum CaSO ₄ ·2H ₂ O/ polymer (PW85)	60%DS gypsum + 20 mg/L PW85	8,453 mg/L	6.7	60	–	–	16.9	Zhao 2006
Collected from sludge holding tank Ballymore Eustace WTP, Southwest Dublin, Ireland	Polymer FO-4140	50 mg/L	2,985 mg/L	6.3	1.16	89	84	–	Ma et al. 2007
Collected from sludge holding tank Ballymore Eustace WTP, Southwest Dublin, Ireland	Polymer LT-25	30 mg/L	2,985 mg/L	6.3	1.16	90	86	–	Ma et al. 2007
Collected from sludge holding tank Ballymore Eustace WTP, Southwest Dublin, Ireland	Dual polymer LT-25/ FO-4140	(1:1) LT-25 15 mg/L + FO-4140 15 mg/L	2,985 mg/L	6.3	1.16	90	84	–	Ma et al. 2007
Collected from underflow channel of sedimentation tank WTP southwest Dublin, Ireland	Fenton (Fe ²⁺ /H ₂ O ₂)	Fe ²⁺ 21 mg/g-DS+H ₂ O ₂ 105 mg g-DS	2850	6.0	1	48	–	–	Tony et al. 2008
Collected from sedimentation tank Ballymore treatment plant Dublin, Ireland	Anionic polymer, Magna-floc LT-25,	3.5 mg/g-DS	2850	6.0	1	67	–	–	Tony et al. 2009
Collected from sedimentation tank Ballymore treatment plant Dublin, Ireland	Cationic polymer, FO-4140 PWG	7.0 mg/g-DS	2850	6.0	1	82	–	–	Tony et al. 2009
Collected from sedimentation tank Ballymore treatment plant Dublin, Ireland	Cu ²⁺ / H ₂ O ₂	Cu ²⁺ 20 mg/g-DS + 125 H ₂ O ₂ mg/g-DS	2850	6.0	1	7	–	–	Tony et al. 2009

Table 2 (continued)

AS, Country of study	Conditioner ^a	Dosage ^b	TSS, mg/L	pH	Tim, min ^b	CST reduc-tion, %	SRF reduc-tion, %	DS increase, %	Ref
Collected from sedimentation tank Ballymore treatment plant Dublin, Ireland	Fenton-like (Fe ³⁺ /H ₂ O ₂)	125 H ₂ O ₂ mg/g DS + Fe ³⁺ 20 mg/g DS	3021	5.7	1	38	64	–	Tony et al. 2011
Collected from WTP, Chongqing, China	Cationic polyacrylamide, CPAM1	8 mg/L	–	7.0	2.5	–	95	32	Sun et al. 2015
Collected from WTP, Chongqing, China	Cationic polyacrylamide CPAM2	16 mg/L	–	7.0	2.5	–	97	55	Sun et al. 2015
Collected from WTP, Chongqing, China	cationic polyacrylamide CPAM3	16 mg/L	–	7.0	2.5	–	96	53	Sun et al. 2015
Collected from WTP, Kedwan, Elminia, Egypt	Solar/Fenton (Fe ²⁺ /H ₂ O ₂)	50mgFeL+800 mg-H ₂ O ₂ /L	2,364	8.5	7	–	78	–	Tony et al. 2016
Collected from WTP, Kedwan, Elminia, Egypt	Solar/anionic polymer, LT-25	10 mg/L	2,364	8.5	3	–	97	–	Tony et al. 2016
Collected from WTP, Kedwan, Elminia, Egypt	Fenton (Fe ²⁺ /H ₂ O ₂)	40mgFe/L + 610 mg-H ₂ O ₂ /L	2,364	7.0	180	–	–	94	Tony and Tayeb 2016

^aCPAM: Poly(acrylamide-acryloyloxyethyl trimethyl ammonium chloride-butylacrylate), PC-320: copolymer of acrylamide and diallyldimethyl-ammonium chloride; CTAB: Cetyltrimethylammonium bromide, SDS: Sodium dodecylsulfate salt, MO: Moringa oleifera seed extract

^bThe dosage and optimal time are shown as the optimal dosage is the investigated range

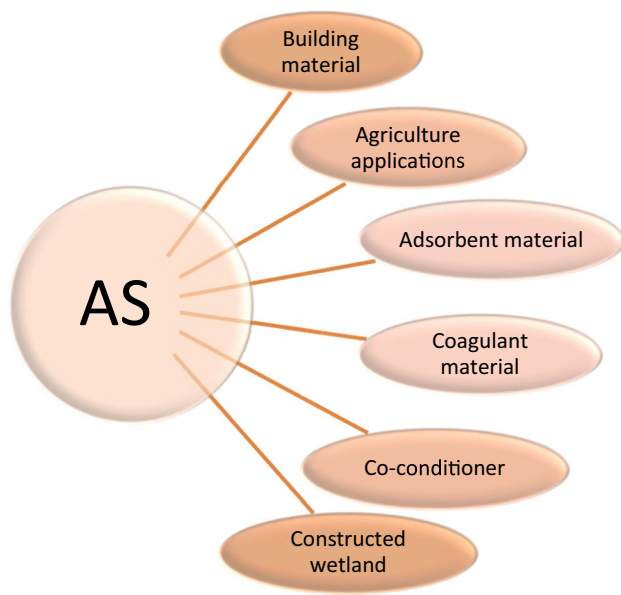


Fig. 6 Summary of the sustainable AS applications

construction substance. However, utilization of alum sludge residues as a building material in the large scale cannot only work as raw material for soil stabilizer, brick manufacturing and cement production, but also could help in its associated waste management problems. For infrastructure and construction, soil stabilization is a vital stage for development.

Traditionally, cement is the most common material used for such job as an additive in a ration of generally used of 12% (Owaid et al. 2014). However, searching for a sustainable environment and waste management opportunities is the scientists continued role. Since the pozzolanic properties of alum are suitable to be a builder material, it could be used as a soil stabilizer as cement or lime (Lin et al. 2005; Bray 2014). In addition, its production is in bulk amounts from the waterworks plants introduces them for such opportunity. Thus, recent literature (Aamir et al. 2019; Henry et al. 2020) is cited dealing with the application of alum sludge as the waterworks residue for soil stabilization as a novel technology. Such application could attain environmentally benign, cost-efficient and sustainable stabilization chance. The data recorded by Aamir et al. (2019) showed an enhancement and improvement in the soil strength–bearing ratio from 6.53 to 16.86% with an optimal alum sludge addition of 8%. However, more research is still required since there is a lack in the literature in such studies. Hence, in the future to studies, it is required to illustrate compaction properties and energy needed to improve soil strength with the merely utilization of the alum sludge waste material.

Cement-based materials

Alum sludge, generally comprised of clay minerals, could be dehydroxylated if heated in the range of 700–850 °C (Tony 2020b). Moreover, the raw waterworks sludge is primarily aluminum-based material with an amorphous aluminum hydroxide precipitate with minimal quantities of crystalline quartz and montmorillonite (Shamaki et al. 2021). The resultant materials are a reactive alumina and silica. Therefore, such materials enhance the pozzolanic reactions when it is supplied to cement improving its characteristics that are related to strength and durability (Fernandez et al. 2011). Thus, calcined alum sludge samples might be signified as a pozzolan since the temperature of calcination has a major influence on the pozzolanic activity (Shamaki et al. 2021). Thus, in cement industry such sludge comprised in past a resultant alite hydration is enhanced since the undersulfated C_3A reactions are prevented; hence, the cement performance is superior.

Sludge recycle is an incinerator in that the cement kiln as a fuel in cement manufacturing is not the only merit and practical alternative, but also it could be as a raw material for cement (Pan et al. 2004). Rodríguez et al. (2010) studied the replacement of 30% of cement with dried alum sludge resulting in a promising reduction in the compressive strength reached 70% in 28 days. Moreover, they reported in their study that dehydration is deduced that is affecting also the setting times of standard mortars. Pan et al. (2004) replaced the clay in cement with clay from fresh alum sludge, which revealed reasonable setting times and the addition did not alter the f-CaO content of the cement. The compressive strength increased with the alum sludge increase and met the standard of the Chinese National Standard of first degree Portland cement. The ternary blends from silica fume, ground-granulated blast furnace slag besides palm oil fuel ash were applied by Owaid and co-workers (2014) with the calcined water works alum sludge at 800 °C. Higher compressive strength was recorded. To add up, Gastaldini et al. (2015) investigated the optimal alum sludge addition in cement industry and their investigation is mainly focused on the optimal calcination temperature which is revealed at 700 °C for one hour of calcination time. In Shamaki and his group (2021) the UK waterworks alum sludge could be utilized with a performance improvement through the addition of gypsum and limestone powder which accelerator effect of calcined sludge dehydration on cement specimen. Thus, the potential use of alum sludge in cement industry has a clear economic driver. Its use as a building material reduces the cost of production.

Brick manufacturing

To meet the bricks demand as building material, clay could be substituted by alum sludge in brick manufacturing. Water works sludge residues based on aluminum waste are mature enough that could be used as a partial replacement of clay in clay brick manufacturing.

Several researchers in many countries used alum sludge in different types of brick making and the compressive strength and shrinkage of the new made brick are explored. For instance, Patricia et al. used waterworks sludge to prepare ceramic brick (Li et al. 2018). Elangovan and Subramanian (2011) introduced the alum sludge with the commercial local clay that is blended in various quantities and sintered at different calcined temperatures to produce clay sludge brick. Also, Tony and Ashour (2014) and Zhao et al. (2016) revealed alum sludge a suitable candidate in clay brick making. Other researchers used water treatment residues as a clay replacement and colorant in facing bricks manufacturing as both colorant material and clay replacement (Andrew and Evaggelia 2007).

Thus, it is supposed from numerous studies that varies types of bricks can be introduced from different countries of studies that meets the regulations of each country by controlling the quantities of incorporated and the sintering temperature. Such research introduces underrated dumped waste alum sludge material in those cases that offer a favorable future for a cost-efficient, economic and environmentally sustainable green options as a promising building materials additive. However, it is notably that the research conducted in using the underrated aluminum-based sludge by-product in construction materials is limited in comparison to other potential applications. This may be related to its organic and water content which makes its chemical composition variable. However, as aforementioned some supplement and treatments to such sludge reveal a suitable geotechnical characteristics and thus geo-environmental applications due to the alteration on its shear strength, mechanical stability and permeability.

Table 3 summarizes the various application of alum sludge (AS) as a building material amendment reported in the previous literature cited. Multiple trials have explored incorporating AS into clay brick (Pan et al. 2004; Ramadan et al. 2008; Santos et al. 2015; Zhao et al. 2016). Initial investigations revealed numerous additions into the brick as well as "AS" such as a combined addition of explored an enhancement in the attained clay brick incinerated sewage sludge ash (Anderson et al. 2003), excavation waste soil (Huang et al. 2005), silica fume and rice husk ash (Hegazy et al. 2012). AS added to Portland cement (Shamsudin et al. 2017) to produce ecological cement brick. Moreover, incorporated limestone (Tony and Ashour 2014) or sand and soil (Shamsudin et al. 2017) with AS into cement to generate

sustainable cement brick showed good compressive strength results.

Preliminary results (displayed in Table 3) of research studies reported the suggested the optimum addition of sludge varied from 5 to 75% to comply the regulations and standard test limits including those in the places of study in UK, India, Egypt, Malaysia, Portugal, Taiwan and Ireland. Investigated outcomes and experimental results from those studies could facilitate the rollout of full-scale factory trials for producing a sustainable brick from AS waste to introduce a green environment opportunity.

It is noteworthy to mention in order to manufacture bricks containing higher amounts of alum sludge, more energy is required. More sintering temperature of the bricks is needed, and the sintering temperature is increased must be increased to ~ 1050–1100 °C. This could be due to the lower silica content and higher water content of alum sludge, which makes the need for more heat. Thus, more intensive energy is required and the process in such case became less economic environmentally appealing (Turner et al. 2019).

Agricultural applications

The applicability of the waterworks sludge containing aluminum is previously applied to the agricultural soils for fertilizing purposes (Dassanayake et al. 2015). Although, it could be applied as a fertilizer in agriculture, the sludge and soil to fertilize must meet some necessities. The aluminum presence in alum sludge could be a source of toxic aluminum and contaminate the environment.

As demonstrated by previous research cited, aluminum and other heavy metals in the sludge could pose a danger to the environment. However, some other researches (Kluczka et al. 2012); (Central Statistical Office Bochenek 2014; Kluczka et al. 2017) contradict this and concluded the soil are not contaminated with heavy metals. Thus, this gets an optimistic vision through the problem and more research could be done for the use of alum sludge in agriculture purposes.

By applying such sludge for agricultural purposes, this introduces the phytotoxic aluminum material into the soil. Such properties are well recognized as harmful to the trees (Ulrich et al. 1980). Other investigator (Smoliński et al. 2009; Yang et al. 2012; Kluczka et al. 2017) reported that the ever increase in aluminum concentrations in soil is signified as one of the main reasons of forests death in North America and Europe. Aluminum not only affects the trees, but also it does cause a certain damage to the crops such as tomatoes, lettuce and beetroot (Brunner and Sperisen 2013). It noteworthy to mention that the elevation in soil acidity notably raises aluminum and heavy metals mobility in the soil (Kluczka et al. 2012). Therefore, the regulations limited the sludge use in the soil to not more than the pH 5.6

Table 3 Concluding research studies on alum sludge application as a construction material*

AS, Country of study	AS properties	AS %	Other additives	Sintering Temp., °C	Application	Potential Outcomes	References
WTP, Pakistan	Al ₂ O ₃ 38%	8	N/A	N/A	Soil	Satisfy Atterberg Limits Tests and Modified Proctor Test; California Bearing Ratio (CBR) Test; Statistical Analysis-Application of ANNs	Aamir et al. 2019
Zhi-Tan WTP northern Taiwan and Feng-Shan plant in southern Taiwan	Mc (18, 56%), Ash content (59, 5%), L.O.I (12, 38%)	N/A	Clay	N/A	Concrete	Compressive strength of the concrete increases with the alum sludge content in the raw meal for cement making without any detrimental effect on long-term strength property	Pan et al. 2004
WTP, Malaysia	600 °C incinerated alum sludge	10	N/A	N/A		The usage of alum sludge in concrete mixes as an alternative of disposal for alum sludge is possible Alum sludge in concrete will directly reduce the quantity of the cement used and decreases CO ₂ emissions	Vasudevan 2019
WTP, India	SiO ₂ 42% Al ₂ O ₃ 35%	15	Fine aggregate (sand), coarse aggregate	N/A		Replacing cement in concrete using alum sludge powder is an effective way of utilizing this chemical and thereby reducing the pollution The difference in percentage weight loss of control mix and alum sludge concrete is less than 2%. So replacing cement by alum sludge powder gave a durable concrete	Suchand et al. 2020
ABBAS water treatment plant in Selangor Malaysia	400 °C treated alum sludge	5	Limestone dust	N/A		Thus, all percentage replacement content showed superior improvement in the concrete workability and said to be a good filling material for concrete	Odimegwu et al. 2020

Table 3 (continued)

AS, Country of study	AS properties	AS %	Other additives	Sintering Temp., °C	Application	Potential Outcomes	References
AS from WWTP oleochemical company, Selangor, Malaysia	MC 70% TOC 15% aluminum sulfate 15%	4	superplasticizer	N/A		Treated AS in concrete is useful -The mechanical strengths of concrete decrease as the replacement level of cement with AS increases The highest reduction in compressive and flexural strengths was up to 66.7%	Ching et al. 2021
Elvington water treatment works located in York, United Kingdom	Alum sludge calcined at 825 C	5	Sand	1100		Calcined sludge added to Portland cement influences cement hydration Calcined Sludge showed the best reactivity due to the formation of poorly crystallized alumina	Shamaki et al. 2021
WTP, United Kingdom	N/A	5	Incinerated sewage sludge ash and clay	N/A	Brick	Well partial replacements for traditional brickmaking raw materials Compressive strength, water absorption or efflorescence, satisfied British Standard kiln emissions	Anderson et al. 2003
Collected from WTP, northern part of Taiwan	pH 6.59 MC 6.27 –11.73%	15	Excavation waste soil	1050		Brick meets the Taiwanese National Science Council's specification for a 1st-grade brick, additions 15–30% produce 2nd–or 3rd–class bricks	Huang et al. 2005
Giza WTP, southern part of Cairo, Egypt	SiO ₂ 43.12%, Fe ₂ O ₃ 5.26%, Al ₂ O ₃ 15.97%	50	Clay	1100		Clay-brick produced superior to the available in the Egyptian market	Ramadan et al. 2008
Bhavani WTP, Erode district, Tamilnadu, India	pH 6.20 Al 12%	20	Clay	950		Satisfies the minimum strength specified in Indian Standard	Elangovan and Subramanian 2011
Giza WTP, Greater Cairo, Egypt	Sludge mainly is quartz & albite	75	Silica fume, rice husk ash, clay	1100		Higher compressive strength compared to 100% clay bricks	Hegazy et al. 2012

Table 3 (continued)

AS, Country of study	AS properties	AS %	Other additives	Sintering Temp., °C	Application	Potential Outcomes	References
Collected from WTP, Kedian, Elminia, Egypt	pH 7.5 MC 97% TSS 2,364 mg/L	5	Limestone, Cement	850		Increasing the sludge amount decreases the compressive strength & increasing water absorption capacity & L.O.I Alum sludge could partial substitute commercial cement bricks without compromising the bricks' strength Additions improved thermal transmittance thus better insulation Thermal transmittance reduction, due to the better thermal performance	Tony and Ashou, 2014
WTP, Portugal	N/A	0–5	Clay	N/A			Santos et al. 2015
WTP ABASS, Malaysia	Al ₂ O ₃ 35%	70	Clay (White Stoneware clay), Silica fume	1150		Fire Clay Brick, the best compressive strength at (Alum in % (70:20:10) of 28.7 MPa & water absorption 11.77% and 13.5%	Odimogwu et al. 2020
Collected WTP Ballymore Eustace Co. Kildare, Dublin, Ireland	pH 6.9 Al 29.3% TSS 2850 mg/L	2	N/A	1200		Brick met the requirement of a load-bearing brick as specified in British Standard Clay Brick met the European and Irish Standards With an increase in sludge content, the color became more pale	Zhao et al. 2016
SAJ Holdings Sdn Bhd WTP, Takzim, Malaysia	N/A	20	Cement, Sand, soil	N/A		Bricks comply with British & Malaysian standards	Shamsudin et al. 2017
Sungai Semenyih WTP, Selangor Malaysia	–	30	Portland cement	1000		Comply the standard of bricks	Firdausand Sobri 2018

N/A not available, MC Moisture content, L.O.I. Loss of Ignition, TOC total Organic Carbon, WWTP wastewater treatment plant

Table 4 Summarized results of alum sludge application in agriculture sector*

AS, Country of study	Plant case study	Other additives	Potential Outcomes	References
WTP Illinois, USA	Corn and soybean	N/A	-No notable effect is re-ordered for the plant nutrients and heavy metals levels in the whole plant parts; thus, AS addition has no detrimental effects on the plants	Lin and Green 1987
WTP, Pennsylvania, USA	Tomato (<i>Lycopersicon esculentum</i>)	Clayey, mixed, mesic typic Ochraquol	-2–10% addition of AS have efficient effect on tomato growth -Increased liming effect, -Reduce Al and Mn toxicity -Reduce heavy metal uptake -alkaline conditions should be avoided	Elliott and Singer 1988
Okla-homa Okla-homa Okla-homa WTP municipalities across Okla-Homa, USA	Tomato (<i>Lycopersicon esculentum</i>)	Other water treatment residuals based polymer (polyaluminum chloride) based	Tomato vegetative yield and tissue P were poor either because of phytotoxic nitrite-nitrogen (NO ₂ -N) (.10 mg/kg) generated during the bioassay or because of WTR P deficiency Limited data suggest that WTRs with NO ₂ -N less than 10 mg/kg and Olsen P greater than 50 mg/kg, water soluble P greater than 580 mg/L, or Mehlich III P greater than 54 mg/kg support growth but still produce inadequate tissue P in tomatoes	Dayton and Basta 2001
Ilsan water purification plant near Seoul, Korea	Indian mustard	Sand Silt Clay	-Increase phosphate adsorption -Reduced leaching of Al, Mg, K, Na & Mn -Increase leaching of Ca -Increase biomass amount and root elongation -Application acid soil is safe & beneficial to plant growth -Require supplemental P fertilization	Kim et al. 2002
WTP, UK	Turf grass	Ferric-based sledges, sub-soil or clay, fertilizers	-The growing of turf is environmentally sustainable and economically attractive using AS -The root action of the grass helping to mineralize the active metal hydroxide -The grass grows well, the root structure is well formed, and the sludge reduces the need for topsoil	Owen 2002

Table 4 (continued)

AS, Country of study	Plant case study	Other additives	Potential Outcomes	References
Howick, WTP, South Africa	Soil improvement	Clay, fine slit	-Increases in hydraulic conductivity and water retention	Moodley et al. 2004
South Africa South Africa WTP, South Africa	Soil enhancements	Sand, silt, clay	-Increases in hydraulic conductivity, water retention and soil water conservation	Moodley and Hughes 2006
Stornoway WT, UK	Soil improvement	Sewage sludge	-Requires evidence of 'agricultural or ecological improvement' -Addition of N, P and K is required	Hastings and Dawson 2012
WTP, Poland	Garden plants	Wastewater sludge	-Allowable limit of EU and Polish Government Regulation are considered and 10% of the total aluminum content of the tested sediments are bioavailable that is danger to the environment	Kluczka et al. 2017

(Kluczka et al. 2017). However, with the ever increasing of the environmental pollution, situation and widespread of the acid rain in the industrial locations recommend monitoring of the bioavailable Al-content is essential (Yang et al. 2012; Kluczka et al. 2017). The summarized results of applying alum sludge as a soil amendment are displayed in Table 4. Dual valuable application of AS into agriculture ameliorant is improving crop production as well as it is an effective sludge management technique. AS can improve soil structure, hydraulic conductivity and nutrient levels. This is related to the presence of organic matter and nutrients in alum sludge. Consequently, alum sludge can be used as a safe soil amendment material to manage and improve soil. However, aluminum toxicity which is present in alum sludge still requires further research to accept AS application in agriculture sector.

Adsorption of pollutants

Due to the chemical nature and amorphous structure of the AS from waterworks residuals impart it with a porous and high surface area material, all of such characteristics introduce it to be a superior adsorbent material (Ippolito et al. 2011; Babatunde and Zhao 2007; Ashour and Tony 2020; Li et al. 2021). Aluminum-based metals could be applied in wastewater treatment sector for numerous contaminants elimination. Various studies (Turner et al. 2019; Tony 2020b) have been investigated the application of alum sludge as a low-cost adsorbent material for contaminants removal from aqueous media. Such contaminants including boron, fluorides, pesticides, perchlorate, glyphosate, dyes

and lead are listed in Table 5. However, detailed and more published studies are related to phosphorus removal from various wastewater effluents and types, their equilibrium isotherm, kinetic analysis and the effect of system parameters have been studied. The presence of amorphous aluminum nature in the Al-based sludge maximizes its affinity to adsorb phosphorous anions from contaminated streams (Babatunde and Zhao 2007; Tony 2021a). AS sorption capabilities are related to the particle size, surface area and surface charge of the AS particles.

Research studies conducted the fully characterization of alum sludge via a complete textural characterization and surface chemistry assessment to signify the surface functional groups and to conduct the adsorption kinetics and equilibrium to explain the adsorption mechanisms (Zhao et al. 2021). The entire adsorption mechanisms include surface precipitation, ligand exchange, electrostatic attraction and ion exchange. It should be noted that although a chemical reaction between pollutants and dissolved aluminum has been demonstrated, it is assumed that the chemical reaction plays only a marginal role in the contaminants removal process (Turner et al. 2019; Liu et al. 2021a, b). The adsorption system could be categorized as physisorption or chemisorption process. In physisorption adsorption, the interaction exists between the solid surface and the adsorbed species has a physical nature in which the interaction forces are depending on the weak van der Waal interaction forces, thus making the process irreversible and requiring low energy barrier. However, chemisorption is signified by the formation of chemical bonds between adsorbed molecules and the solid adsorbent surface via electron transfer or pairing. In contrary

Table 5 Adsorption capacities of numerous uptakes removal from wastewater using AS adsorbents

AS, Country of study	AS (modification/treatment) adsorbent	Adsorbate	Operating conditions	Uptake capacity (mg/g)	References
Lake Major WTP, Dartmouth, Nova Scotia, Canada	Oven-dried	P	AS 4 g/L; Co 4.2 mg-p/L; pH of 6.2; 12 d	0.89 mg/g	Gibbons et al. 2009
Brandon Water Treatment Plant (B-WTP) in Brandon, Manitoba, Canada	Oven-dried	P	AS 5 g; Co. 4.2 mg-p/L; pH 6.8; 12 d	1.59 mg/g	Gibbons et al. 2009
Bradenton, FL, USA	Air-dried, passed through 2-mm sieve	As(V), S(III)	pH 6–6.5; Co 1875 mg/kg; 23 °C; 2 d	7.68 mg/g	Makris et al. 2009
Manatee County Drinking WTP, Bradenton, FL, USA	Air-dried at room temperature (4 weeks), passed through 2-mm screen	Hg	AS 8 mg/kg; pH 6.5; Co 73,816 mg/kg; 7 d	79 mg/g	Hovsepyan and Bonzongo 2009
WTPs in United States and Canada	Oven-dried (105 °C)	As(V)	AS 250 mg/L; pH 8.1; Co 37.7 µg As/L; 20 h	0.001–0.003 mg/g	Gibbons, M. and Gagnon 2010
Tampa, Florida, and Bradenton, Florida, USA	Air-drying, passage through a 2-mm sieve	As(V)	pH 3–7; Co 200 mg/L	12 000 mg/kg	Nagar et al. 2010
WTPs in United States and Canada	Oven-dried (105 °C)	P	pH 6.2; 22 °C; 3.95 mg-p/L	1030–1110 mg/kg	Gibbons and Gagnon 2011
WTPs in United States and Canada	Oven-dried (105 °C)	AsO4	22 °C; pH 6.8; 100 µg AsO4/L	340 mg/kg	Gibbons, M. and Gagnon 2011
Mount Crosby, North Pine WTP Brisbane, Australia	Dried, ground and sieved (<125 µm)	Pb(II)	pH 5–8; 25 °C; 8.5 mg/L	0.21–0.22 mmol/g	Zhou and Haynes (2011)
Mount Crosby, North Pine WTP Brisbane, Australia	Dried, ground and sieved (<125 µm)	Cr(VI)	pH 5–8; 25 °C; 8.5 mg/L	0.37–0.51 mmol/g	Zhou and Haynes 2011
Mount Crosby, North Pine WTP Brisbane, Australia	Dried, ground and sieved (<125 µm)	Cr(III)	pH 5–8; 25 °C; 8.5 mg/L	0.26–0.30 mmol/g	Zhou and Haynes 2011
WTP Abbanoa industry in Truncu Reale, Sassari, Italy	Air-dried, overnight dried (105 °C), grounded	Cd(II) Zn(II)	pH 4.5; 25 °C; 0.1 g; 25 mL; 24 h	0.040 mmol Cd(II)g ⁻¹ 0.050 mmol Zn(II)g ⁻¹	Silvetti et al. 2015
9th WTP, Beijing, China,	Air-dried, ground, sieved (<165 µm)	Co(II)	Co(II) 25–800 mg/L; pH 6.0; 48 h	17.31 mg/g	(Jiao et al. 2017)
Shahid Beheshti WTP, Hamedan, Iran	1 h sedimentation for withdrawing excess water, oven drying	Humic acid	pH 5.56, AS 19.71 g/L, Humic acid 12.28 mg/L	0.47 mg/g	(Foroughi et al. 2018)
water supply plant in Nanjing, China	Air-dried, dried (60 °C), 100-mesh sieve, dried (102 °C), calcination (500 °C, 2 h)	P	AS 0.5 g; pH 7.0; 20 °C; 24 h; 100 mL	0.90 mg/g	Wu et al. 2019
WTP, Shebin El-Kom, Menoufia Governorate, Egypt	Air-dried, oven dried, thermal treatment (400 °C, 2 h)	Phenol	AS 2 g; Co 100–1000 mg L ⁻¹ ; 298 K; 1 h	275 mg-phenol g ⁻¹	Tony 2019
Sludge drying bed of Ming-Der WTP, Miaoli, Taiwan	Dried (105 °C) homogenized for use	Ammonium	AS 300 g; Co 17 g/L; 1000 mL wastewater; 60 min	11.3 mg/g	Cheng et al. 2016
Southern Shebin El-Kom City, Menoufia governorate, Egypt	Gravity settling, air-drying, overnight oven drying, ball mill grinding, calcined (400 °C)	Procion Blue dye	AS 2 g; pH 7; Co 11.8 mg/L; 298 K; 1 h	6.5 mg/g	Tony 2020a, b
Ballymore-Eustace WTP (located in Co. Kildare, South Dublin	Air-dried, grounded, sieved	P	pH 4.3	22.4 mg/g	Zhao and Yang 2010

Table 5 (continued)

AS, Country of study	AS (modification/treatment) adsorbent	Adsorbate	Operating conditions	Uptake capacity (mg/g)	References
Leixlip WTW (located in west Dublin)	Air-dried, grounded, sieved	P	pH 4.3	20.1 mg/g	Zhao and Yang 2010

to the physisorption, chemisorption requires high activation energies and always exists as a monolayer (Tony 2021a, b).

Table 5 summarizes the previous cited studies conducted in the literature using AS as a water works residual material in its nature or modified/treated form for eliminating numerous elements and compounds. As tabulated in the table, the waterworks residuals based on Al-sludge waste could be used and applied for eliminating a wide range of pollutants from wastewater. AS showed a superior effect in phosphorous uptake with high adsorption capabilities is suggested (Gibbons et al. 2009; Gibbons and Gagnon 2010; Wu et al. 2019; Zhao et al. 2021). Their tests with synthetic phosphorous solutions explored that an adsorption capabilities differ according to the operating variables and the initial phosphorous concentration. Additionally, pH has been discovered to have a significant role on the adsorption uptake. The isotherm time could be reached after 24 h (Wu et al. 2019) or 12 days (Gibbons et al. 2009; Gibbons and Gagnon 2011). It could be also applied to remove arsenics from wastewater effluents in a process which is characterized by an increase in the adsorption rate with the increase in the isotherm time which could be reached to 2 days (Makris et al. 2004) at a pH ranged from 3 to 7 according to the type of aqueous effluent (Nagar et al. 2010) (Makris et al. 2009; Gibbons and Gagnon 2010; Nagar et al. 2010). Potentially toxic dye adsorption by AS is well likewise documented in the literature (Tony 2020a, b) for a thermally treated AS samples worked at neutral pH for Procion Blue textile loaded effluents. Scattered studies also showed various pollutants removal from aqueous stream, phenol removal using treated AS has also been investigated (Tony 2019) and Cheng et al. 2016 studied ammonium removal.

Hence, such level of achieved treatment validates the great potential of applying alum sludge (AS) and its environmental benefits, which might be attained through its application especially in developing countries where massive amounts of wastewater from various industries are produced which required to be treated in limited treatment options (Tony and Lin 2021). Such limitations are always related to technical access and cost issues. Otherwise, this AS application is limited to be applied in treating hydrophilic substances. Moreover, the other limitation to AS practical use is the final use of AS after adsorption where they are unsuitable for reuse options and thus need to be landfilled (Wu et al. 2019; Nagar et al. 2010; Turner et al. 2019). It is also noteworthy to mention that the feasibility, cost implications and long-term performances of transforming AS to nanosized particles through milling AS for proper applications could be a reliable option to be a valuable commercially adsorbent for practical applications and reuse facilities (Turner et al. 2019).

Coagulant

Commonly, aluminum coagulants are widely applicable as appropriate coagulant materials to eliminate colloids and phosphorus combinations from water and wastewater systems. In this regard, the most widely applied aluminum coagulant derivatives are aluminum sulfate, sodium tetrahydroxaluminum, aluminum chloride and pre-hydrolyzed aluminum (Tony 2021a, b). While they are not cost-efficient in comparison to ferric coagulants, they are widely applied exclusively in drinking water treatment plants since they have no effect on water quality; thus, this gives its publicity. According to the literature cited, there are published data investigating the impact of the composition and structure of such coagulants to residual aluminum content.

However, such work has only referred to the total Al content and research studies do not introduce the analysis of aluminum fractions (Wolborska et al. 1999; Gregory and Duan 2001; Rak and Świdorska-Bróz, 2001; Yang et al. 2012; Tony and Lin 2020b). In 2010, the regulation of the Minister of Health of Poland regularized 0.2 mg/L as the limited total Al concentration that could be present in the drinking water. Few research studies related to the Al coagulant with its toxic forms could effect in agricultural applications and may cause a danger to soil or not. Results revealed that a larger solubility of aluminum could be achieved when acidification of alum sludge-treated soils to an unamend control soil. Not only acidification could pose a danger, but also, phytotoxicity of soil is related to the pH of the soil (Codling 2008). However, another results found that a negative effect is appeared to the soil plants as a result of alkalinity (Brautigam et al. 2012). For this purpose, the elevation of alum sludge applicability as a coagulant material requires more studies to examine the post-coagulation of the sludge collected from the water treatment plants. Although such use of alum sludge as a coagulant material make it an ideal reuse option, further study is needed. More data are required for full-scale possibilities. However, the viable is still limited, and its future potential use of technology advance can be streamlined.

The combination of alum sludge with ultrafiltration membrane processes has been explored as a promising approach for tertiary wastewater treatment in industrial applications reuses or as a prior treatment technology before the reverse osmosis membrane (Mazari et al. 2018).

Co-conditioner

Nowadays, a considerable attention is gained toward transforming AS waste into a valuable material, rather than an underrated matter for disposal. In this concept, research academia and engineers attain a superior overwhelming research in the last years since AS is most majority by

product from water works plants. Though, actually, limited studies have revealed the use of “AS” as a co-conditioner for sewage sludge dewatering. For example, Lai and Liu (2004) reported that AS acted as a skeleton builder for mixed sludge samples and hence enhanced sludge dewaterability. Yang et al. (2007) and Yang et al. (2009) reported AS application role in condition for improving sewerage sludge dewaterability. Their results demonstrated that polymer dose could be minimized from 120 to 15 mg/L for mixed sludge and thus showed a cost efficient technique. Li et al. (2016) investigated the alum sludge as a drinking water treatment residual which containing polyaluminum chloride and large amount inorganic matters. Chemical conditioner and physical conditioner for improved the sewage sludge dewatering (Tony 2020a). Their results explored that the supplement of AS could reduce the polyacrylamide dosage addition and reduced the moisture content of sewage sludge Ren et al. (2020) demonstrated that alum sludge and sewage sludge mixing in a ratio of 1:1 showed a beneficial AS use as a co-conditioner of sewage sludge and resulted in a good and dewatering results. Realistic economics application of alum sludge co-conditioning opportunity could achieve sustainable reuse development as a conditioner from a waste.

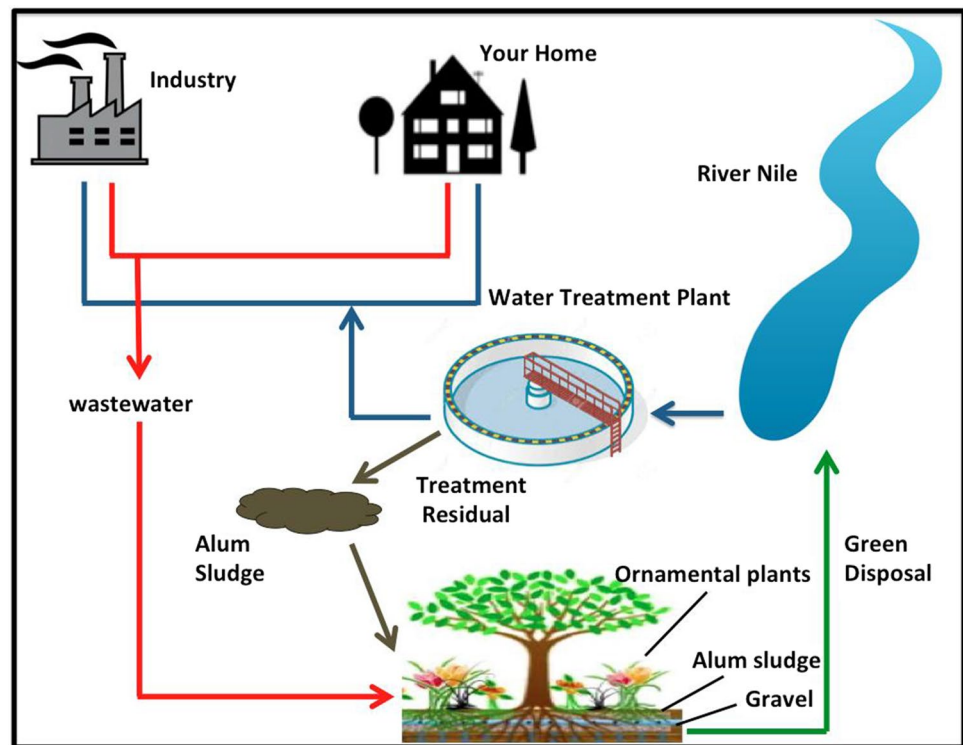
Constructed Wetlands, reed beds and filter beds applications for wastewater treatment

Worldwide, considerable attention has been received in according to the literature published regarding the application of the dewatered AS as a reactive media in constructed wetlands or filter beds. AS is used in the form of AS-cake as the main substrate in constructed wetlands, CW for end of waste lifespan as shown in Fig. 7 (Zhao et al. 2011).

Indeed, substrate is an essential constituent in CW; therefore, the best substrate is chosen that possesses multi-functions, i.e., (i) supporter of biofilm formation; (ii) plants growth media; and (iii) adsorbent for pollutants. From the practical point of view, alum sludge is drinking water treatment plants by-product; hence, it is non-toxic not harmful substance in most cases (Zhao et al. 2013; Turner et al. 2019). Further crucially, aluminum is the predominant constituent in alum sludge which posses' high adsorption affinity to adsorb phosphorus from polluted aqueous streams (Babatunde et al. 2007; Tony 2020b). Hence, aluminum-based sludge cake, as dewatered drinking water treatment plants residuals own a good potential for reuse as raw environmental engineered material as the main substrate in CW for double benefits of reed beds and wastewater treatment.

Extensive studies were explored to assess the capacity and tendency of alum sludge as a low-cost adsorbent material for dyes (Tony 2020a, b), phosphorous (Babatunde et al. 2007; Wu et al. 2019), arsenic (Nagar et al. 2010) and some of heavy metals adsorption (Zhou and Haynes 2011; Castaldi

Fig. 7 Lifespan of alum sludge from source to use in constructed wetlands facilities



et al. 2014; Silveti et al. 2015). Hence, it is introduced as a filter bed or constructed wetland medium (Fig. 7). More research is dealing with its advances only in the last decades, and a focus on the research and development is conducted (Babatunde et al. 2007; Zhao et al. 2008; Hu et al. 2012). The major concern that could face the constructed wetland medium over the long-term applications is clogging. Some studies reported that this issue could be partially alleviated by using anti-sized gravel bed ranged from small at the base of the bed and larger at the top (Zhao et al. 2015). However, Oliver et al. (2011) reported low risk to human health could be attained from organic matter release under anaerobic conditions. Not only using AS as a CW is the solo benefit, but also the final sludge cake after the working lifespan end life could Al and P be recovered. Simple precipitation through pH adjustment is achieved by (Zhao et al. 2013) resulted in 97% and 99% for P and Al, respectively. Hence, practical use of Al-sludge as CW substrate is promising; although the presence of Al for instance still needs further research. Moreover, in the long-term life cycle the clogging issues may still require further development for higher performance yield.

Here, it is important to signify that excellent pollutant removal efficiencies have been cited and well accepted by global constructed wetland research studies. Use of dewatered alum sludge as the main constructed wetland substrate to expand its scope for enhanced wastewater treatment especially phosphorous removal displayed a beneficial reuse of

alum sludge. Developing new materials as substrates in constructed wetland with high adsorption ability and capacity of P or other contaminants is the attaining a priority in wetland advancement.

Al-recovery from AS, impacts and concerns

Viable alternative for end-of-life AS waste is another additional profit besides AS use to reduce the waste produced as well as limiting its final disposal cost. The safe and sustainable alum sludge utilization option in various applications, aluminum recovery could be the reply. Aluminum presence in AS stands is an obstacle from widening its applications. Another potential environmental concern is the probability of elevated level of Al to release to the environment and ecosystem.

Hence, once aluminum is recovered from such sludge, commercial potential of alum sludge utilization in different sectors could be further increased. Al present in AS is about less than 40% by weight. Scattered studies have been previously investigated to remove and recover aluminum from Al-sludge (Dayton and Basta 2001; Vaezi and Batebi 2001; Stendahl et al. 2006).

Numerous progressive procedures are employed to recover aluminum from the aluminum based drinking water residue for converting this environmental issue into a sustainable and economically comprehensive solution. For

instance, some researchers found that Al can be recovered via acid treatments through the pH ranged 1.0–3.0 could acquire Al recovery could achieve Al recovery which could reach to 90% (Abdo et al. 1993; Vaezi and Batebi 2001). Otherwise other researchers reported alkaline treatments at the pH around 11 sodium hydroxide and calcium hydroxide gave a reasonable recovery (Masschelein et al. 1985). Furthermore, membrane separation technology (Sengupta and Shi 1992) and liquid ion exchange methodologies (Cornwell 1982) were also recorded in literature for Al recovery from alum sludge.

Recovery of Al from alum sludge for the option of recycle, reprocess and reuse in order to reduce waste still needs further work data for the full-scale possibilities of coagulant reuse. It is noteworthy to mention that, presently, potential future technologies are required for recovery technologies (Keeley et al. 2014). Till now, according to the current arrangement of recovery data, alum sludge could be recovered and applied in wastewater treatment in less severe regulations regions. No doubt, alum recovering from water treatment residues is minimizing the environmental impacts that could be caused by Al-sludge and also minimize the cost through purchasing alum since the recovered alum may be applied for numerous times (Sanga et al. 2018).

Merits/demerits of sustainable reuse opportunities of 5Rs

Despite the multiple options of alum sludge utilization, still there are unexplored concerns and potential limitations. For instance, in constructed wetland applications, even the disposal route is viable still the lack of the anti-clogging techniques is lack in the literature. The adsorption capabilities of alum sludge are also associated with the pH of the medium. The applicability of AS in soil is also related to the nature of soil, which requires a careful evaluation. Overall, there is still a disconnection between the business enterprise regarding AS applications and the research conducted and academia. Such a lack in connections could be answered through communication of policy makers and scientific support. Otherwise, disposal route such as landfilling of the sludge will be the routine option. Moreover, it is noteworthy to mention that chemical and physical nature of AS differs from one drinking water works plant to another in different countries and regions; thus, further investigation is required before the specific application for definitive conclusion. The merits, demerits and limitations of the diverse alternative AS disposal choices assessed in this article are summarized in Table 6.

Through the 5 Rs' criteria, significant chemical investments and savings are achieved as well as the sludge volume is deduced. Resource recovery and reuse enhance the

treatment efficiency and attain a sustainable environment. But, close distance should be the alum sludge application and handling place and the waterworks plants that produce alum sludge as a sustainable option in order to overcome shipping distances and charges. Some conservation of natural resources could be attained through the utilization of such AS in various applications such as the use in various civil engineering applications in construction industry as a raw material for brick making, concrete and soil amendment. Moreover, heavy metals recovery from sludge and reuse it as coagulant materials or reuse the AS sludge as co-conditioner are also saving the natural resources and sustain our planet. Additionally, agriculture and land sector application of AS is a simple and inexpensive opportunity as a low-grade fertilizer or to attain excess nutrients. Wastewater treatment sector showed diverse pollutants remediation via various AS from different WTPs in different regions.

Generally, controlled application of waterworks residuals "AS" in these abovementioned varies application and feasibly many extra unexplored techniques would valorize alum sludge from "underrated" into "value added" material, in order to maximizing benefits with minimal impact in tandem with the theme of ecological environment.

Concluding glance of development, barriers and future perspectives

More economic alternative for the high disposal costs especially of sludge landfilling is that AS sludge beneficial uses are discussed. Happily, as noted in the above-mentioned applications AS showed various commercial and applications in different regions in the world including UK, Ireland, Europe, the USA, Middle East, Asia and Australia. Particular relevance is given to the reuse in soil applications, building materials, constructed wetland and wastewater treatment. However, still further research is required especially for the field of Al recovery and reuse of alum sludge as a co-conditioner. Cost estimations of alum sludge handling from waterworks plants to the numerous applications require a study and regional regulations tend to the increase of heavy metals recovery options for reuse in agricultural, ecological and industrial purposes.

An updated database in most countries especially in the developing regions is required on quantity and quality of alum sludge by-product to conclude a significant assembly in the present and future AS disposal budgets and controls. Moreover, resource recovery such as heavy metals from alum sludge is considered chemical savings and reduction in sludge volume, but still suffers from operational problems that should be overcome. Further research is still required to prevail long-term constructed wetland drawbacks from any toxic release and long life cycle of anti-clogging systems.

Table 6 Summarized disposal AS alternative of reusing options with their associated merits and impacts*

Application of AS	Merits	Demerits
Building materials	<ul style="list-style-type: none"> ● Nonhazardous ● Reduced shrinkage in clay bricks ● Rich color of the final product ● Organic content may present some energy savings temperature ● Substitute to sand and cementations material ● Best suitable for non-structural concrete ● Metal hydroxide content forms low-porosity brick with acceptable structure ● Nonhazardous ● AS forms calcium aluminum hydrates that helps to prevent chloride corrosion 	<ul style="list-style-type: none"> ● Reduction in compressive strength at higher sludge percentage ● High water absorption ● Requires high sintering temperature ● Organic content in final product may affect mechanical properties ● Risk of hydrogen generation, inclusion of deleterious components ● Aluminum expansion ● Retardation of settling ● Organic content may affect the stability performance ● High water content
Agricultural applications	<ul style="list-style-type: none"> ● Improved soil aggregation ● Increased moisture holding capacity and water permeability ● Effective P reduction at low cost 	<ul style="list-style-type: none"> ● Risk of metal accumulation ● Little fertilizer value ● Potential fixation of available P
Adsorbent	<ul style="list-style-type: none"> ● Suitable for P removal, heavy metal, Textile dye, organic contaminants and pollutants adsorption ● Cost effective ● Possible method for emerging pollutant removals 	<ul style="list-style-type: none"> ● Extent of adsorption is under Research such as organic contaminants remediation ● Possible release of substances ● Risk of clogging ● Does not work well for hydrophilic dyes
CW	<ul style="list-style-type: none"> ● Constructed wetlands substitute wetland media ● Improved P and N removals 	<ul style="list-style-type: none"> ● Transportation distances ● Still need further research
Co-conditioner	<ul style="list-style-type: none"> ● Sludge settling is enhanced ● Sludge dewaterability is improved ● P removal from wastewater 	<ul style="list-style-type: none"> ● Shipping is costive ● Disposal of mixed sludge
Coagulant	<ul style="list-style-type: none"> ● Excellent removal efficiency of turbidity COD and TSS ● Cheap ● Reduction in sludge volume and ● Disposal cost 	<ul style="list-style-type: none"> ● Restricted use due to possibility of contamination ● Shipping cost ● Recovery process ● Limited purity ● Complicated and laborious

COD Chemical Oxygen Demand; TSS Total Suspended Solids

But, for now particular current situation and the probable future trend, there are required attempts for global beneficial reuses of drinking water residues with a critical evaluation for its negative issues from such applications and reuses. Moreover, with increasing attention worldwide to the sustainable and green ecosystem, arising reuse opportunities may be explored.

It is notable that the common use of alum sludge is introduced in adsorption in a powder form after dried and used in grounded and sieved processes. Thus, such powder form of alum sludge stands in the widening of the applications. Moreover, it hinders alum sludge to be attractive adsorbent material since it is difficult to recover after the adsorption process (Zhao et al. 2021). Therefore, in the line of converting alum sludge from a waste into a useful material it is important to granulate it which needs further study.

It is noteworthy to mention that although various reuse applications are present, more work is still needed in the future since the alum sludge material is undervalued material since landfilling is remaining the general final disposal route. Furthermore, regulations concerning the final

discharging of alum sludge and beneficial reuse should be updated. Finally, it is noteworthy to mention that civic belief toward alum sludge applications and reuses and the public anxiety from recycling need urgent believable public education for real applications. Further studies are required for the type of alum sludge introduced for reuse for widening and wiser applications. Hence, a complimentary effort may be necessary for commercial applications in the fields of water, environmental and civil engineering.

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Declarations

Conflict of interest The author declares no potential conflict of interest to report.

Consent for publication The authors confirm the consent for publication.

Human and Animal Rights This article does not contain any studies with human or animal subjects.

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References

- Aamir M, Mahmood Z, Nisar A, Farid A, Ahmed Khan T, Abbas M, Waseem M (2019) Performance evaluation of sustainable soil stabilization process using waste materials. *Processes* 7(6):378
- Abd El-Razek TM, Elgendy AS, Fouad MM (2019) Utilization of drinking water treatment sludge to produce aluminum sulfate. *J Archive*, 48
- Abdo MSE, Ewida KT, Youssef YM (1993) Recovery of alum from wasted sludge produced from water treatment plants. *J Environ Sci Health Part A* 28(6):1205–1216
- Agyin-Birikorang S, O'Connor GA (2009) Aging effects on reactivity of an aluminum-based drinking-water treatment residual as a soil amendment. *Sci Total Environ* 407(2):826–834
- Ahmad T, Ahmad K, Alam M (2016) Sustainable management of water treatment sludge through 3 'R' concept. *J Clean Prod* 124:1–13
- Amaral Filho J, Azevedo A, Etchepare R, Rubio J (2016) Removal of sulfate ions by dissolved air flotation (DAF) following precipitation and flocculation. *Int J Miner Process* 149:1–8
- Anbazzhagan S, Palani S (2018) Extraction of consortium of hydrolytic enzymes from waste activated sludge using ultrasonication and stirring with surfactants. *Ultrason Sonochem* 40:874–880
- Anderson M, Biggs A, Winters C (2003) Use of two blended water industry by-product wastes as a composite substitute for traditional raw materials used in clay brick manufacture. In *Recycling and Reuse of Waste Materials* (pp. 417–426): Thomas Telford Publishing
- Andrew DB, Evaggelia PS (2007) Water treatment residues as clay replacement and colorant in facing bricks. *Characterisation of Mineral Wastes, Resources and Processing technologies—Integrated waste management for the production of construction material-WR0115*
- Ashour EA, Tony MA (2017) Equilibrium and kinetic studies on biosorption of iron (II) and iron (III) ions onto eggshell powder from aqueous solution. *Appl Eng* 1(3):65–73
- Ashour EA, Tony MA (2020) Eco-friendly removal of hexavalent chromium from aqueous solution using natural clay mineral: activation and modification effects. *SN Appl Sci* 2(12):1–13
- Ashour A, Tony MA, Purcell PJ (2014) Use of agriculture-based waste for basic dye sorption from aqueous solution: kinetics and isotherm studies. *Am J Chem Eng* 2(6):92–98
- Babatunde AO, Zhao YQ (2007) Constructive approaches toward water treatment works sludge management: an international review of beneficial reuses. *Crit Rev Environ Sci Technol* 37(2):129–164
- Babatunde AO, Zhao YQ, Yang Y, Kearney P (2007) From "fills" to filter: insights into the reuse of dewatered alum sludge as a filter media in a constructed wetland. *J Residuals Sci Technol* 4(3):147–152
- Basri MHH, Don NNM, Kasmuri N, Hamzah N, Alias S, Azizan FA (2019) Aluminium recovery from water treatment sludge under different dosage of sulphuric acid. *J Phys: Conf Series IOP Publ* 1349:012005
- Bolto B, Gregory J (2007) Organic polyelectrolytes in water treatment. *Water Res* 41(11):2301–2324
- Brautigan DJ, Rengasamy P, Chittleborough DJ (2012) Aluminium speciation and phytotoxicity in alkaline soils. *Plant Soil* 360(1):187–196
- Bray EL (2014) Bauxite and alumina statistics and information. USGS Mineral Industry Surveys.
- Breesem KM, Faris FG, Abdel-Magid IM (2014) Reuse of alum sludge in construction materials and concrete works: a general overview. *Infrastuct Univ Kuala Lumpur Res J* 2(1):20–30
- Brunner I, Sperisen C (2013) Aluminum exclusion and aluminum tolerance in woody plants. *Front Plant Sci* 4:172
- Buyukkamaci N (2004) Biological sludge conditioning by Fenton's reagent. *Process Biochem* 39(11):1503–1506
- Carty G, O'Leary G, Crowe M (2002) *Water treatment manuals: coagulation, flocculation and clarification*. Environ Prot Agency, 85
- Castaldi P, Mele E, Silveti M, Garau G, Deiana S (2014) Water treatment residues as accumulators of oxoanions in soil. sorption of arsenate and phosphate anions from an aqueous solution. *J Hazard Mater* 264:144–152
- Central Statistical Office. (Bochenek, D.- team leader) *Environment 2014 Environment—Information and Statistical Papers* (in Polish), Warsaw
- Cheng W-P, Chen P-H, Yu R-F, Ho W-N (2016) Treating ammonium-rich wastewater with sludge from water treatment plant to produce ammonium alum. *Sustain Environ Res* 26(2):63–69
- Chiang K-Y, Chou P-H, Hua C-R, Chien K-L, Cheeseman C (2009) Lightweight bricks manufactured from water treatment sludge and rice husks. *J Hazard Mater* 171:76–82
- Ching CY, Bashir MJK, Aun NC, Aldahdooh MAA (2021) Sustainable production of concrete with treated alum sludge. *Construct Building Mater* 282:122703
- Christensen JR, Sørensen PB, Christensen GL, Hansen JA (1993) Mechanisms for overdosing in sludge conditioning. *J Environ Eng* 119(1):159–171
- Codling EE (2008) Effects of soil acidity and cropping on solubility of by-product-immobilized phosphorus and extractable aluminum, calcium, and iron from two highphosphorus soils. *Soil Sci* 173:552–559
- Cornwell DA (1982) Process for the extraction of metal ions. In: Google Patents
- Dassanayake KB, Jayasinghe GY, Surapaneni A, Hetherington C (2015) A review on alum sludge reuse with special reference to agricultural applications and future challenges. *Waste Manage* 38:321–335
- Dayton EA, Basta NT (2001) Characterization of drinking water treatment residuals for use as a soil substitute. *Water Environ Res* 73(1):52–57
- De Sena RF, Tambosi JL, Genena AK, de Fpm Moreira R, Schröder HF, José HJ (2009) Treatment of meat industry wastewater using dissolved air flotation and advanced oxidation processes monitored by GC-MS and LC-MS. *Chem Eng J* 152(1):151–157
- Dewil R, Baeyens J, Neyens E (2005) Fenton peroxidation improves the drying performance of waste activated sludge. *J Hazard Mater* 117(2–3):161–170
- Dharmappa HB, Hasia A, Hagare P (1997) Water treatment plant residuals management. *Water Sci Technol* 35(8):45–56

- Elangovan C, Subramanian K (2011) Reuse of alum sludge in clay brick manufacturing. *Water Sci Technol: Water Supply* 11(3):333–341
- Elliott HA, Singer LM (1988) Effect of water treatment sludge on growth and elemental composition of tomato (*Lycopersicon esculentum*) shoots. *Commun Soil Sci Plant Anal* 19(3):345–354
- Fernandez R, Martirena F, Scrivener KL (2011) The origin of the pozzolanic activity of calcined clay minerals: a comparison between kaolinite, illite and montmorillonite. *Cem Concr Res* 41(1):113–122
- Firdaus MS, Sobri S (2018) Performance and evaluation of alum sludge ash as a brick material. Paper presented at the AIP Conference Proceedings
- Foroughi M, Chavoshi S, Bagheri M, Yetilmeszooy K, Samadi MT (2018) Alum-based sludge (AbS) recycling for turbidity removal in drinking water treatment: an insight into statistical, technical, and health-related standpoints. *J Mater Cycles Waste Manage* 20(4):1999–2017
- Förstner U, Haase I (1998) Geochemical demobilization of metallic pollutants in solid waste—implications for arsenic in waterworks sludges. *J Geochem Explor* 62(1–3):29–36
- Foss PJ, O'Connell CA (1996) Irish peatland conservation plan 2000: Irish Peatland Conservation Council Dublin
- Fytily D, Zabaniotou A (2008) Utilization of sewage sludge in EU application of old and new methods—a review. *Renew Sustain Energy Rev* 12(1):116–140
- Gastaldini ALG, Hengen MF, Gastaldini MCC, Do Amaral FD, Antolini MB, Coletto T (2015) The use of water treatment plant sludge ash as a mineral addition. *Constr Build Mater* 94:513–520
- George DB, Berk SG, Adams VD, Ting RS, Roberts RO, Parks LH, Lott RC (1995) Toxicity of alum sludge extracts to a freshwater alga, protozoan, fish, and marine bacterium. *Arch Environ Contam Toxicol* 29(2):149–158
- Ghebremichael KA, Hultman B (2004) Alum sludge dewatering using *Moringa oleifera* as a conditioner. *Water Air Soil Pollut* 158(1):153–167
- Gibbons MK, Gagnon GA (2010) Adsorption of arsenic from a Nova Scotia groundwater onto water treatment residual solids. *Water Res* 44(19):5740–5749
- Gibbons MK, Gagnon GA (2011) Understanding removal of phosphate or arsenate onto water treatment residual solids. *J Hazard Mater* 186(2–3):1916–1923
- Gibbons MK, Mortula MM, Gagnon GA (2009) Phosphorus adsorption on water treatment residual solids. *J Water Supply: Res Technol—AQUA* 58(1):1–10
- Goldman ML, Watson F (1975) Feasibility of alum sludge reclamation: Water Resources Research Center. Washington Technical Institute
- Gregory J, Duan J (2001) Hydrolyzing metal salts as coagulants. *Pure Appl Chem* 73(12):2017–2026
- Guan X-H, Chen G-H, Shang C (2005) Re-use of water treatment works sludge to enhance particulate pollutant removal from sewage. *Water Res* 39(15):3433–3440
- Hastings E, Dawson JJC (2012) The Use of Alum Sludge and Sewage Sludge as a Soil Improver on Barra—Further Response. CREW Publication, Call Down Project (CD2012/16)
- Hegazy BEE, Fouad HA, Hassanain AM (2012) Brick manufacturing from water treatment sludge and rice husk ash. *Aust J Basic Appl Sci* 6(3):453–461
- Henry M, Bauwens T, Hekkert M, Kirchherr J (2020) A typology of circular start-ups: an Analysis of 128 circular business models. *J Clean Prod* 245:118528
- Hidalgo AM, Murcia MD, Gomez M, Gomez E, Garcia-Izquierdo C, Solano C (2017) Possible uses for sludge from drinking water treatment plants. *J Environ Eng* 143(3):04016088
- Hovsepian A, Bonzongo J-CJ (2009) Aluminum drinking water treatment residuals (Al-WTRs) as sorbent for mercury: Implications for soil remediation. *J Hazard Mater* 164(1):73–80
- Hu Y, Zhao Y, Zhao X, Kumar JLG (2012) High rate nitrogen removal in an alum sludge-based intermittent aeration constructed wetland. *Environ Sci Technol* 46(8):4583–4590
- Huang C, Pan JR, Liu Y (2005) Mixing water treatment residual with excavation waste soil in brick and artificial aggregate making. *J Environ Eng* 131(2):272–277
- Ippolito JA, Barbarick KA, Elliott HA (2011) Drinking water treatment residuals: a review of recent uses. *J Environ Qual* 40(1):1–12
- Jiao J, Zhao J, Pei Y (2017) Adsorption of Co (II) from aqueous solutions by water treatment residuals. *J Environ Sci* 52:232–239
- Keeley J, Jarvis P, Judd SJ (2014) Coagulant recovery from water treatment residuals: a review of applicable technologies. *Crit Rev Environ Sci Technol* 44(24):2675–2719
- Keenan HE, Papavasiliopoulos EN, Bache DH (1998) Measurement of polymer residuals in an alum sludge. *Water Res* 32(10):3173–3176
- Kelessidis A, Stasinakis AS (2012) Comparative study of the methods used for treatment and final disposal of sewage sludge in European countries. *Waste Manage* 32(6):1186–1195
- Kim JG, Lee SS, Moon H-S, Kang IM (2002) Land application of alum sludge from water purification plant to acid mineral soil treated with acidic water. *Soil Sci Plant Nutr* 48(1):15–22
- Kluczka J, Żołotajkin M, Ciba J (2012) Speciation of aluminium in the water and bottom sediment of fish-breeding ponds. *Arch Environ Prot*. <https://doi.org/10.2478/v10265-012-0007-7>
- Kluczka J, Żołotajkin M, Ciba J, Staroń M (2017) Assessment of aluminum bioavailability in alum sludge for agricultural utilization. *Environ Monit Assess* 189(8):1–8
- Konieczny K, Szałol D, Płonka J, Rajca M, Bodzek M (2009) Coagulation—ultrafiltration system for river water treatment. *Desalination* 240(1–3):151–159
- Lai JY, Liu JC (2004) Co-conditioning and dewatering of alum sludge and waste activated sludge. *Water Sci Technol* 50(9):41–48
- Li J, Liu L, Liu J, Ma T, Yan A, Ni Y (2016) Effect of adding alum sludge from water treatment plant on sewage sludge dewatering. *J Environ Chem Eng* 4(1):746–752
- Li X, Cui J, Pei Y (2018) Granulation of drinking water treatment residuals as applicable media for phosphorus removal. *J Environ Manage* 213:36–46
- Li Q, Chen Z, Wang H, Yang H, Wen T, Wang S, Hu B, Wang X (2021) Removal of organic compounds by nanoscale zero-valent iron and its composites. *Sci Total Environ* 792:148546
- Lin D-F, Luo H-L, Hsiao D-H, Yang C-C (2005) The effects of sludge ash on the strength of soft subgrade soil. *Sour: J Chin Inst Environ Eng* 15:1–10
- Lin SD, Green CD (1987) A study of wastes from the Centralia water treatment plant and their impact on Crooked Creek. Retrieved from
- Liu F, Hua S, Wang C, Qiu M, Jin L, Hu B (2021) Adsorption and reduction of Cr(VI) from aqueous solution using cost-effective caffeic acid functionalized corn starch. *Chemosphere* 279:130539
- Liu F, Hua S, Wang C, Hu B (2021) Insight into the performance and mechanism of persimmon tannin functionalized waste paper for U(VI) and Cr(VI) removal. *Chemosphere* 287:132199
- Lu MC, Lin CJ, Liao CH, Ting WP, Huang RY (2001) Influence of pH on the dewatering of activated sludge by Fenton's reagent. *Water Sci Technol* 44(10):327–332
- Ma W, Zhao YQ, Kearney P (2007) A study of dual polymer conditioning of aluminum-based drinking water treatment residual. *J Environ Sci Health, Part A* 42(7):961–968
- Makris KC, El-Shall H, Harris WG, O'Connor GA, Obreja TA (2004) Intraparticle phosphorus diffusion in a drinking water

- treatment residual at room temperature. *J Colloid Interface Sci* 277(2):417–423
- Makris KC, Sarkar D, Datta R (2006) Evaluating a drinking-water waste by-product as a novel sorbent for arsenic. *Chemosphere* 64:730–741
- Makris KC, Sarkar D, Parsons JG, Datta R, Gardea-Torresdey JL (2009) X-ray absorption spectroscopy as a tool investigating arsenic (III) and arsenic (V) sorption by an aluminum-based drinking-water treatment residual. *J Hazard Mater* 171(1–3):980–986
- Masschelein WJ, Devleminck R, Genot J (1985) The feasibility of coagulant recycling by alkaline reaction of aluminium hydroxide sludges. *Water Res* 19(11):1363–1368
- Mazari L, Abdessemed D, Szymczyk A (2018) Evaluating reuse of alum sludge as coagulant for tertiary wastewater treatment. *J Environ Eng* 144(12):04018119
- Mikkelsen LH (2001) The shear sensitivity of activated sludge: relations to filterability, rheology and surface chemistry. *Colloids Surf, A* 182(1–3):1–14
- Moodley M, Hughes JC (2006) The effects of a polyacrylamide-derived water treatment residue on the hydraulic conductivity, water retention and evaporation of four contrasting South African soils and implications for land disposal. *Water Sci Technol* 54(5):227–234
- Moodley M, Johnston MA, Hughes JC, Titshall LW (2004) Effects of a water treatment residue, lime, gypsum, and polyacrylamide on the water retention and hydraulic conductivity of two contrasting soils under field conditions in KwaZulu-Natal South Africa. *Soil Res* 42(3):273–282
- Mortula M, Bard SM, Walsh ME, Gagnon GA (2009) Aluminum toxicity and ecological risk assessment of dried alum residual into surface water disposal. *Can J Civ Eng* 36(1):127–136
- Nabarlatz D, Vondrysova J, Jenicek P, Stüber F, Font J, Fortuny A, Bengoa C (2010) Hydrolytic enzymes in activated sludge: extraction of protease and lipase by stirring and ultrasonication. *Ultrason Sonochem* 17(5):923–931
- Nagar R, Sarkar D, Makris KC, Datta R (2010) Effect of solution chemistry on arsenic sorption by Fe- and Al-based drinking-water treatment residuals. *Chemosphere* 78(8):1028–1035
- Odimewu TC, Zakaria I, Abood MM, Kaish A (2020) Workability properties of concrete using alum sludge and limestone dust as replacement of fine aggregate. AIP Publishing LLC, p 020239
- Oliver IW, Grant CD, Murray RS (2011) Assessing effects of aerobic and anaerobic conditions on phosphorus sorption and retention capacity of water treatment residuals. *J Environ Manag* 92:960–966
- Owaid HM, Hamid R, Taha MR (2014) Influence of thermally activated alum sludge ash on the engineering properties of multiple-blended binders concretes. *Constr Build Mater* 61:216–229
- Owen PG (2002) Water-treatment works' sludge management. *Water Environ J* 16(4):282–285
- Pan JR, Huang C, Cherng M, Li K-C, Lin C-F (2003) Correlation between dewatering index and dewatering performance of three mechanical dewatering devices. *Adv Environ Res* 7(3):599–602
- Pan JR, Huang C, Lin S (2004) Reuse of fresh water sludge in cement making. *Water Sci Technol* 50(9):183–188
- Putra RS, Tanaka S (2011) Aluminum drinking water treatment residuals (Al-WTRs) as an entrapping zone for lead in soil by electrokinetic remediation. *Sep Purif Technol* 79(2):208–215
- Rak M, Świdorska-Bróż M (2001) Możliwość minimalizacji niepożądanych skutków procesu koagulacji siarczanem glinu. *Ochrona Środowiska* 3:13–16
- Ramadan MO, Fouad HA, Hassanain AM (2008) Reuse of water treatment plant sludge in brick manufacturing. *J Appl Sci Res* 4(10):1223–1229
- Razali M, Zhao YQ, Bruen M (2007) Effectiveness of a drinking-water treatment sludge in removing different phosphorus species from aqueous solution. *Sep Purif Technol* 55(3):300–306
- Rebosura M Jr, Salehin S, Pikaar I, Kulandaivelu J, Jiang G, Keller J, Yuan Z (2020) Effects of in-sewer dosing of iron-rich drinking water sludge on wastewater collection and treatment systems. *Water Res* 171:115396
- Ren B, Lyczko N, Zhao Y, Nzihou A (2020) Integrating alum sludge with waste-activated sludge in co-conditioning and dewatering: a case study of a city in south France. *Environ Sci Pollut Res* 27(13):14863–14871
- Renou-Wilson F, Moser G, Fallon D, Farrell CA, Müller C, Wilson D (2019) Rewetting degraded peatlands for climate and biodiversity benefits: Results from two raised bogs. *Ecol Eng* 127:547–560
- Rodríguez NH, Ramírez SM, Varela MTB, Guillem M, Puig J, Larrotcha E, Flores J (2010) Re-use of drinking water treatment plant (DWTP) sludge: characterization and technological behaviour of cement mortars with atomized sludge additions. *Cem Concr Res* 40(5):778–786
- Ruhsing Pan J, Huang C, Gang Fu (2000) Effect of surfactant on alum sludge conditioning and dewaterability. *Water Sci Technol* 41(8):17–22
- Sales A, De Souza FR, Almeida FdCR (2011) Mechanical properties of concrete produced with a composite of water treatment sludge and sawdust. *Constr Build Mater* 25(6):2793–2798
- Sanga P, Iradukunda Y, Munyemana JC (2018) Recycling of alum from water treatment residue and reuse it as a flocculating agent for raw water treatment. *J Geosci Environ Protect* 6(12):216
- Santos P, Martins C, Júlio E (2015) Enhancement of the thermal performance of perforated clay brick walls through the addition of industrial nano-crystalline aluminium sludge. *Constr Build Mater* 101:227–238
- Scholz M (2005) Review of recent trends in capillary suction time (CST) dewaterability testing research. *Ind Eng Chem Res* 44(22):8157–8163
- Sengupta AK, Shi B (1992) Selective alum recovery from clarifier sludge. *J Am Water Works Ass* 84(1):96–103
- Shamaki M, Adu-Amankwah S, Black L (2021) Reuse of UK alum water treatment sludge in cement-based materials. *Construct Build Mater* 275:122047
- Shamsudin S-M, Shahidan S, Azmi MAM, Ghaffar SA, Ghani MBA, Bahari NAAS, Zuki SSM (2017) The effect of sludge water treatment plant residuals on the properties of compressed brick. Paper presented at the IOP Conference Series: Materials Science and Engineering
- Sillanpää M, Ncibi MC, Matilainen A, Vepsäläinen M (2018) Removal of natural organic matter in drinking water treatment by coagulation: a comprehensive review. *Chemosphere* 190:54–71
- Silvetti M, Castaldi P, Garau G, Demurtas D, Deiana S (2015) Sorption of cadmium (II) and zinc (II) from aqueous solution by water treatment residuals at different pH values. *Water Air Soil Pollut* 226(9):1–13
- Simpson A, Burgess P, Coleman SJ (2002) The management of potable water treatment sludge: present situation in the UK. *Water Environ J* 16(4):260–263
- Skene TM, Oades JM, Kilmore G (1995) Water treatment sludge: a potential plant growth medium. *Soil Use Manag* 11(1):29–33
- Smoliński A, Zolotajkin M, Ciba J, Dydo P, Kluczka J (2009) PLS-EP algorithm to predict aluminum content in soils of Beskid Mountains region. *Chemosphere* 76(4):565–571
- Sotero-Santos RB, Rocha O, Povinelli J (2005) Evaluation of water treatment sludges toxicity using the *Daphnia* bioassay. *Water Res* 39(16):3909–3917
- Stendahl K, Färm C, Fritzdorf I, Ulmert H (2006) The REAL process—a process for recycling sludge from water works. *Water Sci Technol* 54(5):235–242

- Suchand SS, Dr Leena AV (2020) Alum sludge -a partial replacement to cement in concrete. *Inter. Res J Eng Technol* 7(7):0056–0072
- Sun Y, Fan W, Zheng H, Zhang Y, Li F, Chen W (2015) Evaluation of dewatering performance and fractal characteristics of alum sludge. *PLoS one* 10(6):e0130683
- Tayeb AM, Tony MA, Ismael EK (2019) Engineered nanostructured ZnO for water remediation: operational parameters effect, Box-Behnken design optimization and kinetic determinations. *Appl Water Sci* 9(3):1–11
- Teh CY, Budiman PM, Shak KPY, Wu TY (2016) Recent advancement of coagulation–flocculation and its application in wastewater treatment. *Ind Eng Chem Res* 55(16):4363–4389
- Tetteh EK, Rathilal S (2019) Application of organic coagulants in water and wastewater treatment. *Org. Polym*
- Thabet RH, Tony MA, El Sherbiny SA, Ali IA, Fouad MK (2020) Catalytic oxidation over nanostructured heterogeneous process as an effective tool for environmental remediation. *IOP Conf Series: Mater Sci Eng* 975:012004
- Thabet RH, Fouad MK, Ali IA, El Sherbiny SA, Tony MA (2021) Magnetite-based nanoparticles as an efficient hybrid heterogeneous adsorption/oxidation process for reactive textile dye removal from wastewater matrix. *Int J Environ Anal Chem*, 1–23
- Thomas B, Murphy DJ, Murray BG (2016) *Encyclopedia of applied plant sciences*. Academic Press
- Tony MA (2020a) Central composite design optimization of Bismarck Dye oxidation from textile effluent with Fenton's reagent. *Appl Water Sci* 10(5):1–9
- Tony MA (2020b) Zeolite-based adsorbent from alum sludge residue for textile wastewater treatment. *Int J Environ Sci Technol*. <https://doi.org/10.1007/s13762-020-02646-8>
- Tony MA (2021) Low-cost adsorbents for environmental pollution control: a concise systematic review from the prospective of principles, mechanism and their applications. *J Dispers Sci Technol*. <https://doi.org/10.1080/01932691.2021.1878037>
- Tony MA (2021) Nexus approach: ZSM12 derived from industrial waste and organic wax pair for microencapsulated solar energy storage system, energy sources, part a recovery, utilization, and environmental effects. *Energy Sour Part A*. <https://doi.org/10.1080/15567036.2021.2001119>
- Tony MA, Ali AI (2021) Mechanistic implications of redox cycles solar reactions of recyclable layered double hydroxides nanoparticles for remazol brilliant abatement. *Int J Environ Sci Technol*. <https://doi.org/10.1007/s13762-021-03818-w>
- Tony MA, Ashour E (2014) Sustainable development: Effects of reusing water treatment plant sludge in brick manufacturing, accepted for oral presentation in the 7th international conference for chemical and environmental engineering, Military technical college, May 27–29. Egypt, Cairo
- Tony MA, Lin L-S (2020) Attenuation of organics contamination in polymers processing effluent using iron-based sludge: process optimization and oxidation mechanism. *Environ Technol*. <https://doi.org/10.1080/09593330.2020.1803417>
- Tony MA, Lin LS (2020b) Performance of acid mine drainage sludge as an innovative catalytic oxidation source for treating vehicle-washing wastewater. *J Dispersion Sci Technol*. <https://doi.org/10.1080/01932691.2020.1813592>
- Tony MA, Lin L-S (2021) Iron coated-sand from acid mine drainage waste for being a catalytic oxidant towards municipal wastewater remediation. *Int J Environ Res*. <https://doi.org/10.1007/s41742-020-00309-7>
- Tony MA, Tayeb AA (2016) Response surface regression model in optimization of alum sludge drying facility: solar-fenton's reagent dewatering. *Int J Chem Eng Appl* 7(5):331
- Tony MA, Zhao YQ, Fu JF, Tayeb AM (2008) Conditioning of aluminium-based water treatment sludge with Fenton's reagent: effectiveness and optimising study to improve dewaterability. *Chemosphere* 72(4):673–677
- Tony MA, Zhao YQ, Tayeb AM (2009) Exploitation of Fenton and Fenton-like reagents as alternative conditioners for alum sludge conditioning. *J Environ Sci* 21(1):101–105
- Tony MA, Zhao YQ, El-Sherbiny MF (2011) Fenton and fenton-like aops for alum sludge conditioning: effectiveness comparison with different Fe²⁺ and Fe³⁺ salts. *Chem Eng Commun* 198(3):442–452. <https://doi.org/10.1080/00986445.2010.520235>
- Tony MA, Tayeb A, Zhao YQ (2016) An alternative arrangement for the alum sludge management: minimising waste with low-cost solar techniques. *Am J Chem Eng* 4(2):30–37
- Tony M (2019) Win-win wastewater treatment to sustain world: Porous adsorbents from waste waterworks sludge for phenol remediation. Paper presented at the Anaerobic Digestion Conference AD16, The International water Association, IWA
- Turner T, Wheeler R, Stone A, Oliver I (2019) Potential alternative reuse pathways for water treatment residuals: remaining barriers and questions—a review. *Water Air Soil Pollut* 230(9):1–30
- Ulén B, Etana A, Lindström B (2012) Effects of aluminium water treatment residuals, used as a soil amendment to control phosphorus mobility in agricultural soils. *Water Sci Technol* 65(11):1903–1911
- Ulrich B, Mayer R, Khanna PK (1980) Chemical changes due to acid precipitation in a loess-derived soil in central Europe. *Soil Sci* 130(4):193–199
- Vaezi F, Batebi F (2001) Recovery of iron coagulants from Tehran water-treatment-plant sludge for reusing in textile wastewater treatment
- Vasudevan G (2019) Performance of Alum Sludge as partial replacement for cement adding superplasticizer. Paper presented at the IOP Conference Series: Materials Science and Engineering
- Wang D, He Y (2010) Status and Source Analysis of Aluminum Pollution in Xi'an Section of the Weihe River. Paper presented at the 4th International Conference on Bioinformatics and Biomedical Engineering
- WHO (2011) *WHO Guidelines for Drinking-Water Quality*. 4th ed
- Wolborska A, Zarzycki R, Mętlewiak B (1999) Influence of the type of aluminum coagulant on treated water quality. *Ochrona Środowiska* 4:25
- Wu CC, Wu JJ, Huang RY (2003) Floc strength and dewatering efficiency of alum sludge. *Adv Environ Res* 7(3):617–621
- Wu HF, Wang JP, Duan EG, Feng YF, Wan ZY, Wu YX, Lu YQ (2019) Study on the preparation of granular alum sludge adsorbent for phosphorus removal. *Water Sci Technol* 79(12):2378–2386
- Xiong JB, Mahmood Q (2010) Adsorptive removal of phosphate from aqueous media by peat. *Desalination* 259(1–3):59–64
- Yang Y, Tomlinson D, Kennedy S, Zhao YQ (2006) Dewatered alum sludge: a potential adsorbent for phosphorus removal. *Water Sci Technol* 54(5):207–213. <https://doi.org/10.2166/wst.2006.564>
- Yang Y, Zhao YQ, Babatunde AO, Kearney P (2007) Co-conditioning of the anaerobic digested sludge of a municipal wastewater treatment plant with alum sludge: benefit of phosphorus reduction in reject water. *Water Environ Res* 79(13):2468–2476
- Yang Y, Zhao YQ, Babatunde AO, Kearney P (2009) Two strategies for phosphorus removal from reject water of municipal wastewater treatment plant using alum sludge. *Water Sci Technol* 60(12):3181–3188
- Yang Z, Gao B, Wang Y, Zhao Y, Yue Q (2012) Fractionation of residual Al in natural water treatment from reservoir with poly-aluminum-silicate-chloride (PASiC): effect of OH/Al, Si/Al molar ratios and initial pH. *J Environ Sci* 24(11):1908–1916
- Yin C-Y (2010) Emerging usage of plant-based coagulants for water and wastewater treatment. *Process Biochem* 45(9):1437–1444

- Yin X, Han P, Lu X, Wang Y (2004) A review on the dewaterability of bio-sludge and ultrasound pretreatment. *Ultrason Sonochem* 11(6):337–348
- Yu L, Han M, He F (2017) A review of treating oily wastewater. *Arab J Chem* 10:S1913–S1922
- Yu S, Pang H, Huang S, Tang H, Wang S, Qiu M, Chen Z, Yang H, Song G, Fu D, Hu B, Wang X (2021) Recent advances in metal-organic framework membranes for water treatment: a review. *Sci Total Environ* 800:149662
- Zhang S, Wang J, Zhang Y, Ma J, Huang L, Shujun Y, Chen L, Song G, Qiu M, Wang X (2021) Applications of water-stable metal-organic frameworks in the removal of water pollutants: a review. *Environmental Pollution* 291:118076
- Zhao YQ (2004) Settling behaviour of polymer flocculated water-treatment sludge I: analyses of settling curves. *Sep Purif Technol* 35(1):71–80
- Zhao YQ (2006) Involvement of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) in water treatment sludge dewatering: a potential benefit in disposal and reuse. *Sep Sci Technol* 41(12):2785–2794
- Zhao YQ, Bache DH (2001) Conditioning of alum sludge with polymer and gypsum. *Colloids Surf, A* 194(1–3):213–220
- Zhao YQ, Bache DH (2002) Integrated effects of applied pressure, time, and polymer doses on alum sludge dewatering behaviour. *Waste Manage* 22(7):813–819
- Zhao Y, Yang Y (2010) Extending the use of dewatered alum sludge as a P-trapping material in effluent purification: study on two separate water treatment sludges. *J Environ Sci Health Part A* 45:1234–1239
- Zhao YQ, Babatunde A, Razali M, Harty F (2008) Use of dewatered alum sludge as a substrate in reed bed treatment systems for wastewater treatment. *J Environ Sci Health - Part A Toxic/Hazard Subst Environ Eng* 43(1):105–110
- Zhao YQ, Keogh C, Tony MA (2009) On the necessity of sludge conditioning with non-organic polymer: AOP approach. *J Residuals Sci Technol* 6(3):151–155
- Zhao XH, Hu YS, Kumar JU, Babantunde OA, Zhao YQ (2011) Pilot field-scale demonstration of a novel alum sludge-based constructed wetland system for enhanced wastewater treatment. *Process Biochem* 46(1):278–283
- Zhao XH, Zhao YQ, Kearney P (2013) Phosphorus recovery as AlPO₄ from beneficially reused aluminium sludge arising from water treatment. *Environ Technol* 34(2):263–268
- Zhao X, Zhao Y, Wang W, Yang Y, Babatunde A, Hu Y, Kumar L (2015) Key issues to consider when using alum sludge as substrate in constructed wetland. *Water Sci Technol* 71:1775–1782
- Zhao Y, Ren B, O'Brien A, O'Toole S (2016) Using alum sludge for clay brick: an Irish investigation. *Int J Environ Stud* 73(5):719–730
- Zhao Y, Liu R, Awe OW, Yang Y, Shen C (2018) Acceptability of land application of alum-based water treatment residuals—an explicit and comprehensive review. *Chem Eng J* 353:717–726
- Zhao W, Xie H, Li J, Zhang L, Zhao Y (2021) Application of alum sludge in wastewater treatment processes: “science” of reuse and reclamation pathways. *Processes* 9(4):612
- Zhou Y-F, Haynes RJ (2011) Removal of Pb (II), Cr (III) and Cr (VI) from aqueous solutions using alum-derived water treatment sludge. *Water Air Soil Pollut* 215(1):631–643
- Zhou X, Jiang G, Wang Q, Yuan Z (2014) A review on sludge conditioning by sludge pre-treatment with a focus on advanced oxidation. *RSC Adv* 4(92):50644–50652
- Zhu Y, He X, Xu J, Fu Z, Wu S, Ni J, Hu S (2021) Insight into efficient removal of Cr(VI) by magnetite immobilized with *Lysinibacillus* sp. JLT12: Mechanism and performance. *Chemosphere* 262:127901

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