



# Alum sludge as an adsorbent for inorganic and organic pollutants removal from aqueous solutions: a review

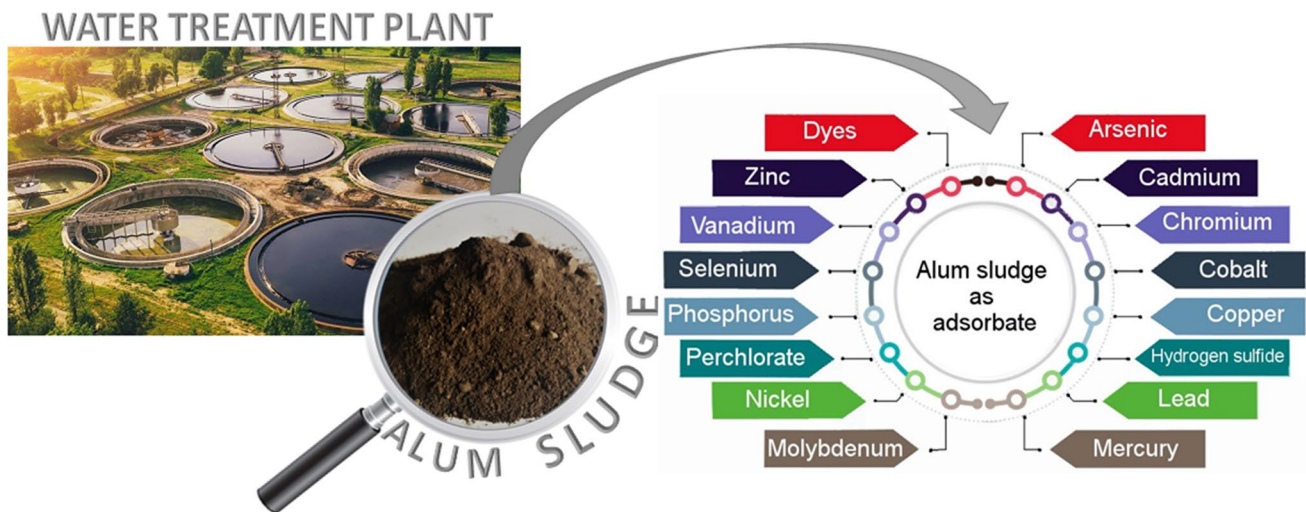
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Received: 30 November 2022 / Revised: 14 February 2023 / Accepted: 18 February 2023 / Published online: 9 March 2023  
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## Abstract

One of the first stages of water treatment is the coagulation process, a side effect of which is the formation of huge amounts of sludge, which is most often deposited in landfills. The residues formed as a by-product in water treatment plants are characterized by high porosity, irregularity and loose structure with limited strength, so they can exhibit adsorption properties. This article reviews the ability of Al-based sludge to adsorb inorganic pollutants and textile dyes from aqueous solutions. Due to its physicochemical properties, alum sludge has a high adsorption capacity against inorganic as well as organic pollutants. The collected literature review focuses on the presentation of the most common directions of use of these materials as effective adsorbents against the pollutants present, the presentation of the conditions for the adsorption process, the mechanisms of binding of pollutants. In addition, it indicates which directions of their use as effective and economical adsorbents have not been sufficiently explored, suggesting that more research in this area be conducted.

## Graphical abstract



**Keywords** Adsorption · Alum sludge · Drinking water waste · Dyes · Heavy metals · Inorganic pollutants · Water treatment residues

Editorial responsibility: Xiao-Zhang Yu.

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## Introduction

Water—a basic good for some, a luxury good for others. According to the World Health Organization, around 2.1 billion people worldwide do not have access to drinking

water at home, and more than twice as many are unsanitary. This means that 1 in 3 people do not have access to drinking water, and 6 in 10 people do not have basic sanitation. According to estimates by the United Nations, the availability of clean and safe drinking water may drop by as much as 40% in the next decade. By 2050, water demand will double, and more than half of the world's population will be at risk of water shortages (World Water Assessment Programme (Nations Unies), The United Nations World Water Development Report 2018 (United Nations Educational, Scientific and Cultural Organization, New York, USA) [www.unwater.org/publications/world-water-development-report-2018/](http://www.unwater.org/publications/world-water-development-report-2018/) (2018)).

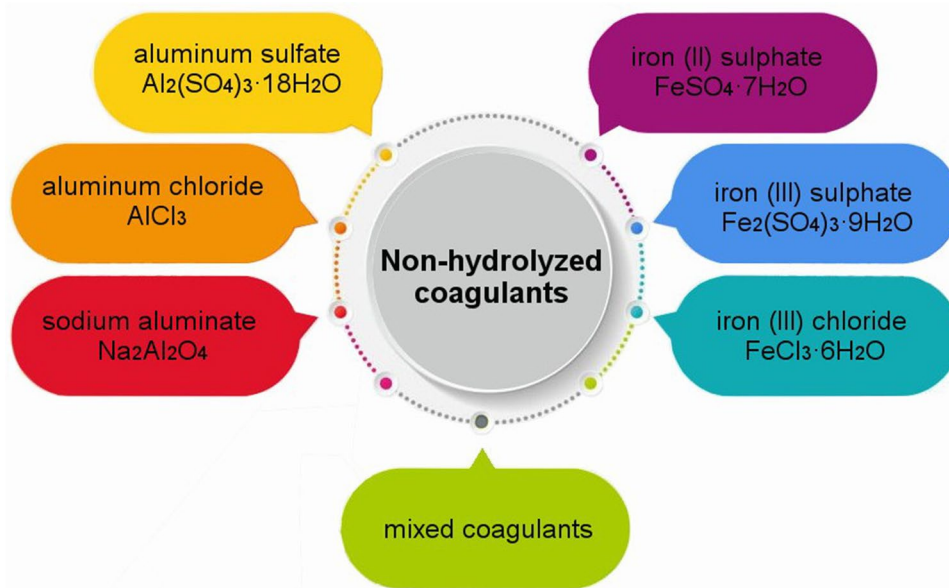
Fresh water is becoming the most critical resource problem facing humanity. While fresh water supply is limited, both the world's population and demand for fresh water continue to grow rapidly. The world's rapid population growth over the last century, the constantly growing needs of society, the development of civilization with the simultaneous increase in awareness of the quality of drinking water and, as a consequence, the tightening of regulations concerning pollution of the aquatic environment by the European Union Directives force the creation of more effective, ecologically safe, and because of the constantly increasing water prices (resulting from the costs of extraction, water treatment and more expensive wastewater treatment processes) also of economical methods for water and wastewater treatment (Boretti and Rosa 2019).

One of the first stages of water treatment is the coagulation, which is a well-established process in water treatment, to remove suspended particles by combining small particles into larger aggregates. Coagulation is a process to neutralize charges and then to form a gelatinous mass to trap (or

bridge) particles thus forming a mass large enough to settle or be trapped in the filter. The effect produced by the addition of a chemical to a colloidal dispersion results in particle destabilization by the reduction in the forces tending to keep the particles apart (Dassanayake et al. 2015; Hargreaves et al. 2018; Ippolito et al. 2009, 2011; Nourmoradi et al. 2016; Oliver et al. 2011; Zhou and Haynes 2011). This process unfortunately has the disadvantage of producing huge amounts (typically 10–30 mL of sludge for every liter of water treated) of sludge-like waste belonging to water treatment residue (WTR) (Liu et al. 2016; Yang et al. 2008). It is estimated that conventional wastewater treatment plants produce about 100,000 tons of sludge per year, although due to the ever-increasing demand for treated water, it can be assumed that this amount may continue to rise. Therefore, waste management requires appropriate directions that are both economical and environmentally friendly (Liu et al. 2016; Turner et al. 2019; Yang et al. 2008).

The main components of WTR are separated soils, organic matter and hydrated metal oxides of Al and Fe, depending on the metal salt used for coagulation (Guan et al. 2005; Hou et al. 2018; Ippolito et al. 2011; Keeley et al. 2014). Coagulants based on hydrolysable metal salts such as aluminum and iron are widely used (Nagar et al. 2010; Nair and Mansoor Ahammed 2015; Ong et al. 2017; Scalize et al. 2019; Zhou and Haynes 2010). Metal salts quickly hydrolyze to form different types of cations that are adsorbed by negatively charged particles and cause charge reduction (Hargreaves et al. 2018; Liu et al. 2016). The coagulation process uses two types of coagulants, non-hydrolyzed aluminum and iron salts and pre-hydrolyzed ones. The most commonly used non-hydrolyzed coagulants are shown in Fig. 1.

**Fig. 1** Non-hydrolyzed coagulants (Dassanayake et al. 2015; Ippolito et al. 2009; Nourmoradi et al. 2016; Oliver et al. 2011; Zhou and Haynes 2011)



The most frequently used coagulant in drinking water treatment is aluminum sulfate  $Al_2(SO_4)_3$  (Babatunde et al. 2008; Guan et al. 2005; Jeon et al. 2018; Liu et al. 2016; Qi et al. 2011; Yang et al. 2006a, 2008; Zhao et al. 2009).

According to Barakwan et al. (2019), Hou et al. (2018), Keeley et al. (2014), alum sludge (AS) is most often land-filled, which is an important environmental problem, to which not enough attention is paid. In addition, the AS may be used to:

- Protect against water and wind erosion of the escarpment and crown surface of a closed landfill (Babatunde and Zhao 2007; Balkaya 2015),
- For liming acid soils (Kim et al. 2002),
- As a soil ameliorant (Dayton and Basta 2005; Lombi et al. 2010),
- For reusing as a coagulant for the treatment of urban wastewater (Ahmad et al. 2016; Guan et al. 2005; Nair and Mansoor Ahammed 2015; Zhao et al. 2009, 2011),
- As a co-conditioner in sewage sludge conditioning and dewatering process (Babatunde and Zhao 2007; Yang et al. 2006a),

- As a constructed wetland substrate (Babatunde et al. 2008),
- For the production of cement mortar (Chi-Liang et al. 2011; Yu-Chi et al. 2012),
- For the production of bricks (Babatunde and Zhao 2007; Chiang et al. 2012; Zhao et al. 2009, 2011; Yu-Chi et al. 2012),
- For the production of tiles and ceramic pipes (Babatunde and Zhao 2007; Jordán et al. 2005),
- For the glass–ceramic production (Tarrago et al. 2017),
- For agricultural use (Dassanayake et al. 2015) (Fig. 2).

Contemporary sludge management in water treatment plants is understood as the whole of activities that enable them to: introduction into the environment or recultivated in a way safe, compliant with applicable regulations, storage at landfills, use in the economy or industry, use in wastewater treatment systems through the use of recovered from reactant sediments or by recycling recycled water supernatant, disposal with sewage sludge or waste (Hou et al. 2018; Keeley et al. 2014). However, as a balanced approach to mitigating these effects, current

**Fig. 2** Various directions of use and management of AS (Ahmad et al. 2016; Babatunde and Zhao 2007; Babatunde et al. 2008; Balkaya 2015; Dassanayake et al. 2015; Guan et al. 2005; Kim et al. 2002; Lombi et al. 2010; Nair and Mansoor Ahammed 2015; Tarrago et al. 2017; Yang et al. 2006a; Zhao et al. 2009, 2011)



trends indicate a gradual move toward the reuse of alumina sludge as a beneficial material. Such beneficial reuse includes the use of AS as an adsorbent to remove pollutants from water and wastewater (Bai et al. 2014; Barakwan et al. 2019; Caporale et al. 2013; Geng et al. 2018; Ghorpade and Mansoor Ahammed 2018; Hovsepyan and Bonzongo 2009; Jiao et al. 2017; Jo et al. 2021; Nagar et al. 2010; Ong et al. 2017; Quinones et al. 2016; Ren et al. 2021; Zhao et al. 2021). It is well known adsorption due to the simplicity of construction, profitability, efficiency, ease of use, tolerance to toxic materials, biodegradability is a more promising method of treating water and wastewater from inorganic and organic pollutants (Crini 2006; Gupta and Suhas 2009; Ippolito et al. 2011). And the best known and most widely used adsorbent is activated carbon (Crini 2006; Gupta and Suhas 2009; Pająk 2021). However, the high cost of this material forces the search for new low-cost, effective, unconventional adsorbents, such as natural and waste materials for water and wastewater treatment. Such promising material is AS.

Many scientific publications on the disposal of post-coagulation sludge are available (Gadekar and Mansoor Ahammed 2020; Genc-Fuhrman et al. 2007; Hou et al. 2018; Ippolito et al. 2009; Jeon et al. 2018; Jo et al. 2021; Makris et al. 2006; Md Nor et al. 2014; Nourmoradi et al. 2016; Poormand et al. 2017; Ren et al. 2021; Wang et al. 2016; Wołowiec et al. 2019; Zhao et al. 2011; Zhou and Haynes 2011); however, the availability of review articles on the adsorption properties of post-coagulation sludge and its applicability for the removal of inorganic and organic pollutants from water and wastewater are significantly limited and mainly concern the reuse of sludge in various fields, not only as adsorptive materials (Babatunde and Zhao 2007; Dassanayake et al. 2015; Ippolito et al. 2011; Johnson et al. 2014; Turner et al. 2019; Wołowiec et al. 2019; Zhao et al. 2018, 2021).

In view of the above, the review will focus on the following aspects: formation and characteristics of post-coagulation sludge, physicochemical properties of AS, adsorption properties of AS in relation to various pollutants, pollutant binding mechanism, adsorption isotherms.

It was observed that the post-coagulation sludge formed in water treatment plants by the authors is referred to variously, namely as AS, water treatment residues (WRT), alum-derived water treatment sludge (Al-WRT), alum-based drinking water treatment residues (Al-based DWTR) or drinking water waste (DWW). However, all the results presented the adsorption capacity of Al-based sludge, where aluminum salts, variously named by different

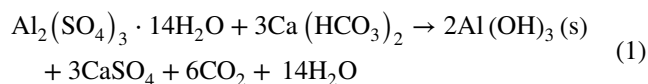
authors, were used as coagulant (Ahmad et al. 2016; Babatunde et al. 2008; Bai et al. 2014; Gadekar and Mansoor Ahammed 2020; Hua et al. 2018; Ippolito et al. 2009; Ong et al. 2017).

3rd January, 2022–30th November, 2022, Institute of Environmental Engineering Polish Academy of Science, Poland.

## Materials and methods

### Composition and characteristics of the AS

AS is a by-product of drinking water treatment in water treatment plants where aluminum salt is used as a coagulant. AS is an aggregate of organic matter, humic particles and colloidal suspended solids, and its main component  $\text{Al}(\text{OH})_3$  is amorphous in nature and is formed when the  $\text{Al}_2(\text{SO}_4)_3$  coagulant reacts with hydroxides ( $\text{OH}^-$ ) when added to water at a sufficiently high concentration as shown in Eq. 1 (Hua et al. 2018; Ippolito et al. 2011; Jo et al. 2021; Kim et al. 2012; Kumar et al. 2020):



The amount, composition and properties of AS strongly depend on the quality and quantity of the treated water, the type and dose of coagulants used (aluminum salts used in the treatment of surface waters and iron salts used in the treatment of groundwater) and the process conditions (mainly mixing). Therefore, depending on the above factors, the physicochemical properties of the AS differ from each other, which is presented in Table 1.

AS is characterized by high porosity, irregularity and loose structure with limited strength, so that they can exhibit adsorptive properties (Awab et al. 2012; Babatunde et al. 2009; Liu et al. 2016). The researchers observed that the specific surface area of AS can range from 21 to  $364.55 \text{ m}^2 \text{ g}^{-1}$ , which is about 1/3 of the surface of activated carbon (Table 1). Zhou and Haynes (2010), while studying the adsorption capacity of AS for heavy metals ions, observed that the specific surface area of AS is much higher than that of other adsorptive materials such as coal fly ash or bauxite processing residue mud (red mud). Ren et al. (2021) in their research on the use of AS as an efficient adsorbent for the removal of hydrogen sulfide showed also that AS has a specific surface area greater than “normal” adsorbents, such as charred wood pellets, fly ash from



**Table 1** Physicochemical characteristic of the AS

Parameter/properties	Unit	Range	References
SSA	m <sup>2</sup> g <sup>-1</sup>	21–435.5	Awab et al. (2012), Babatunde et al. (2009), Bai et al. (2014), Caporale et al. (2013), Castaldi et al. (2014), Hovsepian and Bonzongo (2009), Hua et al. (2018), Jo et al. (2021), Kim et al. (2012), Makris et al. (2006), Nagar et al. (2010), Silveti et al. (2015), Siswoyo et al. (2014), Soleha et al. (2016), Zhou and Haynes (2010, 2011, 2012)
Porosity	%	41–69	Awab et al. (2012), Babatunde et al. (2009), Liu et al. (2016)
CEC	cmol kg <sup>-1</sup>	6.2–66.76	Hovsepian and Bonzongo (2009), Hua et al. (2018), Ippolito et al. (2009), Silveti et al. (2015), Zhou and Haynes (2011, 2012)
EC	mS cm <sup>-1</sup>	0.36–6.9	Hovsepian and Bonzongo (2009), Ippolito et al. (2003, 2009), Nagar et al. (2010), Silveti et al. (2015), Zhou and Haynes (2010, 2012)
pH	–	4.28–7.90	Ahmad et al. (2016), Awab et al. (2012), Bai et al. (2014), Barakwan et al. (2019), Butani and Mane (2017), Castaldi et al. (2014), Gadekar and Mansoor Ahammed (2016), Ghorpade and Mansoor Ahammed (2018), Ippolito et al. (2009), Rashed et al. (2016), Scalize et al. (2019), Silveti et al. (2015), Siswoyo et al. (2014), Zhou and Haynes (2011, 2012)
pH <sub>PZC</sub>	–	5.6–6.9	Ghorpade and Mansoor Ahammed (2018), Hua et al. (2018), Silveti et al. (2015), Zhou and Haynes (2010, 2011, 2012)
Organic matter	%	12–23.17	Hou et al. (2018), Lee et al. (2015), Nagar et al. (2010), Silveti et al. (2015)
Moisture content	%	2.35–95	Ahmad et al. (2016), Awab et al. (2012), Hou et al. (2018), Lee et al. (2015)
Solid content	%	2.4–12.24	Ahmad et al. (2016), Butani and Mane (2017), Gadekar and Mansoor Ahammed (2016), Guan et al. (2005), Nair and Mansoor Ahammed (2015)
Ash	%	35.12–85	Awab et al. (2012), Hou et al. (2018), Silveti et al. (2015), Siswoyo et al. (2014)
Mineral compositions	Alumina (Al <sub>2</sub> O <sub>3</sub> ) Gibbsite (Al(OH) <sub>3</sub> ) Calcite (CaCO <sub>3</sub> ) Magnesite (MgCO <sub>3</sub> ) α-Goethite (FeO(OH)) Kaolinite (Al <sub>2</sub> H <sub>4</sub> O <sub>9</sub> Si <sub>2</sub> ) Quartz (SiO <sub>2</sub> ) Hematite (Fe <sub>2</sub> O <sub>3</sub> ) Gehlenite (Ca <sub>2</sub> Al <sub>2</sub> SiO <sub>7</sub> ) Amorphous material	–	Balkaya (2015), Georgantas and Grigoropoulou (2005), Kim et al. (2002), Soleha et al. (2016), Zhou and Haynes (2010, 2012)
<i>Chemical composition</i>			
Al	mg g <sup>-1</sup>	12.80–174.6	Awab et al. (2012), Babatunde and Zhao (2010), Babatunde et al. (2009), Butani and Mane (2017), Gadekar and Mansoor Ahammed (2016, 2020), Ghorpade and Mansoor Ahammed (2018), Hou et al. (2018), Hovsepian and Bonzongo (2009), Ippolito et al. (2003, 2009), Krishna et al. (2016), Makris et al. (2005, 2006), Nair and Mansoor Ahammed (2015), Siswoyo et al. (2014), Silveti et al. (2015), Zhou and Haynes (2010, 2011, 2012)
Fe	mg g <sup>-1</sup>	3.336–90.0	Awab et al. (2012), Babatunde and Zhao (2010), Babatunde et al. (2009), Butani and Mane (2017), Gadekar and Mansoor Ahammed (2016, 2020), Ghorpade and Mansoor Ahammed (2018), Hou et al. (2018), Hovsepian and Bonzongo (2009), Ippolito et al. (2003, 2009), Krishna et al. (2016), Makris et al. (2005, 2006), Nair and Mansoor Ahammed (2015), Silveti et al. (2015), Siswoyo et al. (2014), Zhou and Haynes (2010, 2011, 2012)
Ca	mg g <sup>-1</sup>	0.081–129.83	Bai et al. (2014), Butani and Mane (2017), Gadekar and Mansoor Ahammed (2016, 2020), Ghorpade and Mansoor Ahammed (2018), Zhou and Haynes (2012)
C	mg g <sup>-1</sup>	12.7–243	Bai et al. (2014), Caporale et al. (2013), Hovsepian and Bonzongo (2009), Nagar et al. (2010), Zhou and Haynes (2010, 2011)



**Table 1** (continued)

Parameter/properties	Unit	Range	References
Si	mg g <sup>-1</sup>	3.82–123.5	Ghorpade and Mansoor Ahammed (2018), Hua et al. (2018), Nair and Mansoor Ahammed (2015), Zhou and Haynes (2012)
Mg	mg g <sup>-1</sup>	0.091–49.16	Butani and Mane (2017), Gadekar and Mansoor Ahammed (2016, 2020), Ghorpade and Mansoor Ahammed (2018), Hou et al. (2018), Maqbool et al. (2015), Nair and Mansoor Ahammed (2015)
K	mg g <sup>-1</sup>	0.00383–20.06	Ahmad et al. (2016), Butani and Mane (2017), Gadekar and Mansoor Ahammed (2016, 2020), Ghorpade and Mansoor Ahammed (2018), Hou et al. (2018), Ippolito et al. (2003), Krishna et al. (2016), Silvetti et al. (2015), Zhou and Haynes (2011)
N	mg g <sup>-1</sup>	1.06–8.68	Caporale et al. (2013), Hovsepian and Bonzongo (2009), Ippolito et al. (2003), Nagar et al. (2010), Silvetti et al. (2015)
Cu	mg g <sup>-1</sup>	0.0171–4.11	Awab et al. (2012), Hovsepian and Bonzongo (2009), Ippolito et al. (2003, 2009), Siswoyo et al. (2014)
P	mg g <sup>-1</sup>	0.89–3.9	Ahmad et al. (2016), Babatunde et al. (2009), Bai et al. (2014), Caporale et al. (2013), Ghorpade and Mansoor Ahammed (2018), Ippolito et al. (2003), Makris et al. (2006), Silvetti et al. (2015)
Mn	mg g <sup>-1</sup>	0.030–2.2	Awab et al. (2012), Babatunde et al. (2009), Ghorpade and Mansoor Ahammed (2018), Ippolito et al. (2009), Krishna et al. (2016), Silvetti et al. (2015), Siswoyo et al. (2014)
Cd	mg g <sup>-1</sup>	0.00177–1.6	Awab et al. (2012), Ippolito et al. (2003), Poormand et al. (2017)
Na	mg g <sup>-1</sup>	0.1565–1.6	Ghorpade and Mansoor Ahammed (2018), Hua et al. (2018), Krishna et al. (2016), Silvetti et al. (2015), Zhou and Haynes (2012)
Ba	mg g <sup>-1</sup>	0.048–0.49	Ippolito et al. (2009), Krishna et al. (2016)
Cr	mg g <sup>-1</sup>	0.0143–0.0811	Awab et al. (2012), Hovsepian and Bonzongo (2009), Ippolito et al. (2003, 2009)
Pb	mg g <sup>-1</sup>	0.00199–0.0371	Awab et al. (2012), Babatunde and Zhao (2010), Babatunde et al. (2009), Hovsepian and Bonzongo (2009), Ippolito et al. (2003)
As	mg g <sup>-1</sup>	0.00801–0.034	Babatunde and Zhao (2010), Babatunde et al. (2009), Caporale et al. (2013), Hovsepian and Bonzongo (2009), Ippolito et al. (2003), Siswoyo et al. (2014)
Ni	mg g <sup>-1</sup>	0.0066–0.0125	Awab et al. (2012, Ippolito et al. (2003, 2009)
Hg	mg g <sup>-1</sup>	0.00002–0.00066	Hovsepian and Bonzongo (2009), Ippolito et al. (2003)
B	mg g <sup>-1</sup>	0.0916	Ippolito et al. (2009)
Sr	mg g <sup>-1</sup>	0.0311	Ippolito et al. (2009)
Mo	mg g <sup>-1</sup>	0.0037	Ippolito et al. (2003)
Se	mg g <sup>-1</sup>	0.0001–0.0015	Ippolito et al. (2003, 2009)
Al <sub>2</sub> O <sub>3</sub>	%	12.1–51.01	Ahmad et al. (2016), Balkaya (2015), Rashed et al. (2016), Razali et al. (2007), Zhao et al. (2007)
Fe <sub>2</sub> O <sub>3</sub>	%	0.70–10.32	Ahmad et al. (2016), Balkaya (2015), Rashed et al. (2016), Razali et al. (2007), Zhao et al. (2007)
CaO	%	1.2–4.39	Ahmad et al. (2016), Balkaya (2015), Razali et al. (2007), Zhao et al. (2007)
SiO <sub>2</sub>	%	1.6–54.72	Ahmad et al. (2016), Balkaya (2015), Rashed et al. (2016)
MgO	%	0.22–3.08	Ahmad et al. (2016), Balkaya (2015)
Na <sub>2</sub> O	%	0.07–0.97	Ahmad et al. (2016), Balkaya (2015)
K <sub>2</sub> O	%	0.20–3.62	Ahmad et al. (2016), Balkaya (2015)
TiO <sub>2</sub>	%	0.05–20.65	Ahmad et al. (2016), Balkaya (2015)
P <sub>2</sub> O <sub>5</sub>	%	0.17–0.36	Ahmad et al. (2016), Balkaya (2015)
MnO	%	0.08–0.49	Ahmad et al. (2016), Balkaya (2015)
ZnO	%	0.01	Ahmad et al. (2016)

municipal waste incineration or rice husk-activated carbon. Awab et al. (2012), Hua et al. (2018), Jo et al. (2021), Soleha et al. (2016) explain the high specific surface area by the

amorphous nature of the AS using the Scanning electron microscopy analysis (SEM). Awab et al. (2012) on the basis of the appearance of the SEM and field emission scanning



electron microscopy analysis (FESEM) conclude that the AS is amorphous in nature, and in order to determine the components or minerals in the samples, they additionally carried out an *X-ray diffraction* analysis (XRD), so that a qualitative description of the sample sediment structure can also be provided. Gadekar and Mansoor Ahammed (2020) on the basis of the SEM show the amorphous nature of AS, obtained as a result of aggregation, rearrangement and hydrolysis to amorphous hydroxide precipitate. Also, Soleha et al. (2016) noted that the micrographs clearly show that the surface of the AS was rough and porous; therefore, they are good candidates as adsorbents.

Large surface area as a positive attribute of potential adsorbents was recognized by Zhou and Haynes (2012). These researchers, based on X-ray diffraction analysis, showed that the AS consists of poorly crystalline amorphous material, although small amounts of gehlenite were also present. Additionally, they observed that when alum dissolves in water,  $\text{Al}^{3+}$  exists in aqueous solution as aluminohexahydrone ions  $[\text{Al}(\text{H}_2\text{O})_6]^{3+}$  and they dissociate sequentially with increasing pH (leaving  $\text{OH}^-$  instead of  $\text{H}_2\text{O}$ ) to form compounds such as  $[\text{Al}(\text{H}_2\text{O})_5\text{OH}]^{2+}$  and  $[\text{Al}(\text{H}_2\text{O})_4(\text{OH})_2]^+$ , which then polymerize to form positively charged multinuclear complexes. Zhou and Haynes (2010) argue that adsorption of inorganic and organic anionic compounds to hydroxy-Al polymers inhibits their crystallization to  $\text{Al}(\text{OH})_3$ , resulting in a relatively stable amorphous material. Also, other researchers (Bai et al. 2014; Jo et al. 2021; Silveti et al. 2015), on the basis of XRD or SEM analyses of the AS, showed that these materials have the amorphous nature.

The values of the cation exchange capacity (CEC), electrical conductivity (EC), values of pH and point of zero charge ( $\text{pH}_{\text{PZC}}$ ) of the AS differ significantly. The lowest CEC values were reported by Zhou and Haynes (2011) during their study on the use of AS for the removal of Pb(II), Cr(III) and Cr(VI) from water, while the highest values were reported by Silveti et al. (2015) during their study on the adsorption of Cd(II) and Zn(II) from aqueous solution by AS. Also, the EC values of AS were in a wide range and varied among different sludges. The pH values of the AS varied in a wide range depending on the tested AS, while the value of the  $\text{pH}_{\text{PZC}}$  was usually at a similar level for most researchers in the range of 5.6–6.9 (Table 1).

Moreover, it was observed that the content of organic matter (OM) in the AS differed depending on the tested sludge, and the lowest value content was reported by Lee et al. (2015) in a study on the reuse of AS from a local water treatment plant in Singapore using aluminum sulfate

as coagulant, while the highest OM contents were reported by Silveti et al. (2015). The AS was characterized by a different value of the solid content. The lowest value of the solid content was recorded during the study on the re-use of AS to enhance particulate pollutant removal from sewage (Guan et al. 2005). These researchers argue the low solids content by the fact that the AS had not gone through any thickening and/or dewatering processes. AS with the highest solid content was studied by Butani and Mane (2017) in their work on coagulation/flocculation processes for the removal of cationic and anionic dyes using AS, which were collected from a water treatment plant (Nigdi, Pune), where polyaluminum chloride (PAC) is used in the coagulation/flocculation process.

AS also differed in the ash content. The lowest values of ash content were recorded by Siswoyo et al. (2014) during the study of AS collected from Miyamachi and Nishino DWTP in the city of Sapporo, Hokkaido, Japan, as an effective and low-cost adsorbent for the removal of Cd ions in water. The highest ash in the research was obtained by Hou et al. (2018), while investigating the characteristics and maximum adsorption capacity of dehydrated AS for P from four water treatment plant in China's Shandong Province.

AS consisted of alumina (Babatunde et al. 2008; Balkaya 2015; Rashed et al. 2016; Razali et al. 2007; Soleha et al. 2016; Yang et al. 2006b; Zhao et al. 2007), and other minerals such as gibbsite, calcite, magnesite,  $\alpha$ -goethite, kaolinite, mica, quartz, hematite, gehlenite (Balkaya 2015; Georgantas and Grigoropoulou 2005; Kim et al. 2002; Soleha et al. 2016; Zhou and Haynes 2010, 2012). The mineral composition of the AS is reflected in the chemical composition which is presented in Table 1.

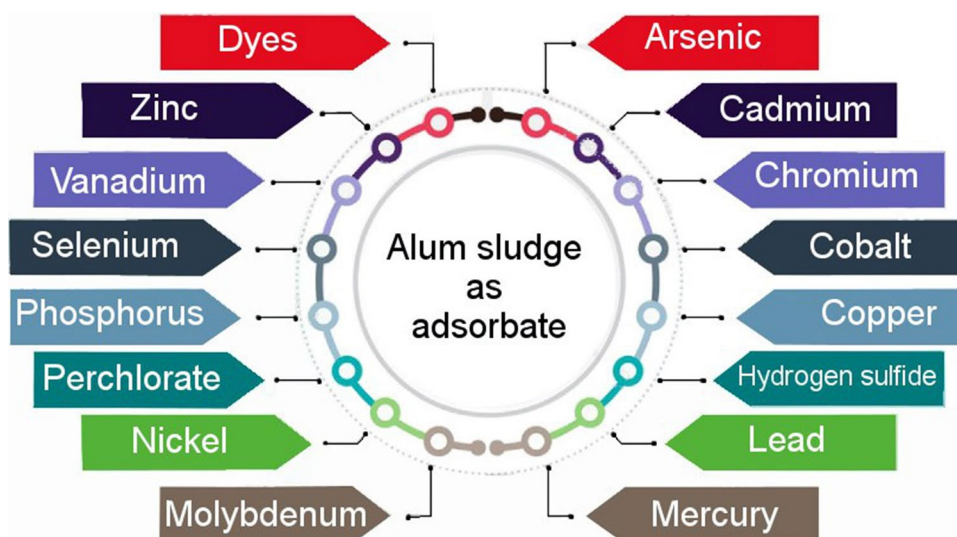
## Results and discussion

### Adsorption of pollutants using AS

The studies of the properties of AS in the largest amounts concern the removal of phosphorus (P) (Babatunde et al. 2008; Bai et al. 2014; Dayton and Basta 2005; Georgantas and Grigoropoulou 2005; Gibbons and Gagnon 2011; Hou et al. 2018; Krishna et al. 2016; Maqbool et al. 2015; Razali et al. 2007; Wang et al. 2016; Yang et al. 2006a, 2006b, 2008; Zhao and Yang 2010; Zhao et al. 2007). P is an essential nutrient for the growth of aquatic plants, but too much P compounds in the water lead to eutrophication of the aquatic environment. A large amount of nutrient compounds causes a massive bloom of algae and cyanobacteria, which in turn



**Fig. 3** Pollutants adsorbed on AS (Babatunde et al. 2008; Caporale et al. 2013; Ghorpade and Mansoor Ahammed 2018; Hovsepyan and Bonzongo 2009; Hua et al. 2015; Jeon et al. 2018; Jiao et al. 2017; Kim et al. 2012; Lian et al. 2020; Md Nor et al. 2014; Rashed et al. 2016; Silveti et al. 2015; Siswoyo et al. 2014; Wang et al. 2016; Yang et al. 2008; Yusuff et al. 2017; Zhou and Haynes 2010, 2011, 2012)



adversely affects water quality by reducing oxygen content, and this leads to the death of aquatic organisms. P is found in typical wastewater, both domestic and industrial, and occurs mainly as phosphate in dissolved or particulate form. The phosphates can be inorganic and condensed phosphates or organic form (Babatunde et al. 2008; Wang et al. 2016; Yang et al. 2008).

AS is an effective adsorbent for removing P from water and wastewater (Babatunde et al. 2008; Bai et al. 2014; Hou et al. 2018). The P-adsorption capacity strongly depends on the pH of the solution, where as pH increases, the surface charge of the AS changes from positive to negative (Babatunde et al. 2008; Yang et al. 2006a, 2008). However, AS can also be an effective adsorbent for removing heavy metals ions, inorganic pollutants and textile dyes from water and wastewater. The adsorption capacity of the AS for various pollutants presented in Fig. 3 was investigated.

#### Adsorption of inorganic pollutants using AS

In the face of more and more stringent regulations, pollutants such as As, Se, V, Mo, Cd, Zn, Pb, Cr, Cu, Co, Hg are now priority pollutants for the environment and become one of the most serious environmental problems. The problem is compounded by the fact that these pollutants are commonly found in soil, water, wastewater in excessive concentrations. Using of AS in removal of these pollutants is the object of widespread interest in the world. Table 2 shows adsorption capacity, removal efficiency and experimental conditions of AS for various pollutants from aqueous solutions.

**As, Se, V and Mo adsorption onto AS** As and Se are listed in the United States Environmental Protection Agency (US-EPA) priority pollutant list with known adverse and toxic effects on environment and human health (Chiang et al. 2012; Elkhatib et al. 2015; Genc-Fuhrman et al. 2007; Hua et al. 2018; Ippolito et al. 2009; Jo et al. 2021; Kim et al. 2020; Makris et al. 2007; Tugrul et al. 2013). The most common oxidation numbers of As are (+5), (+3) and (−3), in which the element is able to form both inorganic (more toxic) and organic compounds in the environment (Caporale et al. 2013), while Se can exist in different oxidation states, elemental selenium (Se), selenite ( $\text{SeO}_3^{2-}$ ), selenide ( $\text{Se}^{2-}$ ), selenate ( $\text{SeO}_4^{2-}$ ) and organic selenium in the environment, but under ordinary alkaline and oxidized conditions the predominant form is selenate (Zhou and Haynes 2012). Moreover, V and Mo are often discharged together with As from various industries related to the manufacture of alloy steels.

Several authors have extensively studied the potential use of AS as an effective adsorbent for As removal from aqueous solutions. Among others, Jeon et al. (2018) conducted a study of As adsorption on AS modified by calcination, resulting in decomposition of natural organic matter, subsequent crystallization of aluminum mineral structure and consequent increase in the adsorption capacity of the AS. The researchers further observed that As(V) adsorbed onto AS could be desorbed by alkaline solution and the adsorbent could be reused as an adsorbent for As(V). The researchers noted that the heterogeneity of AS and its surface properties played an important role in the adsorption of As(V), where various adsorption sites and heterogeneous composition of minerals on AS had different affinities to As(V).



**Table 2** Adsorption capacity, removal efficiency and experimental conditions of AS for various pollutants from aqueous solutions

Adsorbate	Adsorption capacity ( $q_{max}$ )	Conditions for adsorption process	Kinetic model	Isotherm	Adsorption mechanism	Thermo-dynamic	References
As (V)	$q_{max} = 22.10 \text{ mg g}^{-1}$	$m = 0.1 \text{ g}$ , $C_0 = 100 \text{ mg L}^{-1}$ , $\text{pH} = 6.0$ , $T = 50 \text{ }^\circ\text{C}$ , $t = 24 \text{ h}$	Pseudo-I-order Pseudo-II-order	Freundlich	Chemisorption Diffusion	Endothermic	Jeon et al. (2018)
	$q_{max} = 15,000 \text{ mg kg}^{-1}$	$m = 1 \text{ g}$ , $\text{pH} = 6.0$ , $T = 23 \text{ }^\circ\text{C}$ , $t = 48 \text{ h}$	Pseudo-II-order	Freundlich	Diffusion	-	Makris et al. (2007)
	$q_{max} = 7.68 \text{ mg g}^{-1}$	$m = 1 \text{ g}$ , $C_0 = 100 \text{ mg L}^{-1}$ , $\text{pH} = 4.0$ , $t = 20 \text{ }^\circ\text{C}$ , $T = 49 \text{ h}$	Pseudo-II-order	Freundlich	Diffusion	Endothermic	Kim et al. (2012)
	$q_{max} = 0.28 \text{ mmol g}^{-1}$	$m = 10 \text{ g}$ , $C_0 = 6 \text{ mM}$ , $\text{pH} = 5.0$ , $T = 25 \text{ }^\circ\text{C}$ , $t = 2 \text{ h}$	Pseudo-II-order	Langmuir	Electrostatic repulsion	-	Zhou and Haynes (2012)
As (III)	$q_{max} = 600.0\text{--}667.1 \text{ mmol kg}^{-1}$	$m = 5 \text{ g}$ , $C_0 = 0.125\text{--}6.25 \text{ mmol L}^{-1}$ , $\text{pH} = 3.0\text{--}9.0$ , $T = 20 \text{ }^\circ\text{C}$ , $t = 24 \text{ h}$	-	Langmuir	Chemisorption	-	Caporale et al. (2013)
	$q_{max} = 17.36 \text{ mg g}^{-1}$	$m = 10 \text{ g}$ , $C_0 = 1 \text{ mg L}^{-1}$ , $\text{pH} = 3.0\text{--}10.0$ , $T = 25 \text{ }^\circ\text{C}$ , $t = 2 \text{ h}$	Pseudo-II-order	Freundlich Langmuir	Chemisorption	-	Hua et al. (2015)
	$q_{max} = 8900 \text{ mg kg}^{-1}$	$m = 1 \text{ g}$ , $\text{pH} = 6.0$ , $T = 23 \text{ }^\circ\text{C}$ , $t = 48 \text{ h}$	Pseudo-II-order	Freundlich	Diffusion	-	Makris et al. (2007)
	$q_{max} = 16.53 \text{ mg g}^{-1}$	$m = 1 \text{ g}$ , $C_0 = 100 \text{ mg L}^{-1}$ , $\text{pH} = 4.0$ , $T = 20 \text{ }^\circ\text{C}$ , $t = 49 \text{ h}$	Pseudo-II-order	Langmuir	Diffusion	Exothermic	Kim et al. (2012)
Se (VI)	$q_{max} = 0.25 \text{ mmol g}^{-1}$	$m = 10 \text{ g}$ , $C_0 = 6 \text{ mM}$ , $\text{pH} = 5.0$ , $T = 25 \text{ }^\circ\text{C}$ , $t = 2 \text{ h}$	Pseudo-II-order	Freundlich	Electrostatic repulsion	-	Zhou and Haynes (2012)
	$q_{max} = 459.1\text{--}537.1 \text{ mmol kg}^{-1}$	$m = 5 \text{ g}$ , $C_0 = 0.125\text{--}6.25 \text{ mmol L}^{-1}$ , $\text{pH} = 3.0\text{--}9.0$ , $T = 20 \text{ }^\circ\text{C}$ , $t = 24 \text{ h}$	-	Langmuir	Chemisorption	-	Caporale et al. (2013)
	$q_{max} = 0.14 \text{ mmol g}^{-1}$	$m = 10 \text{ g}$ , $C_0 = 1 \text{ mM}$ , $\text{pH} = 2.0\text{--}12.0$ , $T = 25 \text{ }^\circ\text{C}$ , $t = 2 \text{ h}$	Pseudo-II-order	Freundlich	Electrostatic repulsion	-	Zhou and Haynes (2012)
	$q_{max} = 1400\text{--}2100 \text{ mg kg}^{-1}$	$C_0 = 60 \text{ mg L}^{-1}$ , $\text{pH} = 5.0\text{--}9.0$ , $T = 25 \text{ }^\circ\text{C}$ , $t = 24 \text{ h}$	-	-	Chemisorption (Inner sphere complexation)	-	Ippolito et al. (2009)
Se (IV)	$q_{max} = 0.28 \text{ mmol g}^{-1}$	$m = 10 \text{ g}$ , $C_0 = 1 \text{ mM}$ , $\text{pH} = 2.0\text{--}12.0$ , $T = 25 \text{ }^\circ\text{C}$ , $t = 2 \text{ h}$	Pseudo-II-order	Langmuir	Electrostatic repulsion	-	Zhou and Haynes (2012)

Table 2 (continued)

Adsorbate	Adsorption capacity ( $q_{\max}$ )	Conditions for adsorption process	Kinetic model	Isotherm	Adsorption mechanism	Thermo-dynamic	References
Mo (VI)	$q_{\max} = 1400\text{--}1950 \text{ mg kg}^{-1}$ $q_{\max} = 39.52 \text{ mg g}^{-1}$	$C_0 = 60 \text{ mg L}^{-1}$ , pH = 5.0–9.0, $T = 25^\circ\text{C}$ , $t = 24 \text{ h}$ $m = 1 \text{ g}$ , $C_0 = 0\text{--}100 \text{ mg L}^{-1}$ , pH = 4.0, $T = 25^\circ\text{C}$ , $t = 24 \text{ h}$	– Pseudo-II-order	– Langmuir Temkin Dubinin-Radushkevich	Chemisorption (Inner sphere complexation) Physisorption Chemisorption	– Endothermic	Ippolito et al. (2009) Lian et al. (2020)
V (V)	$q_{\max} = 9.53 \text{ mg g}^{-1}$	$m = 10 \text{ g}$ , $C_0 = 1 \text{ mg L}^{-1}$ , pH = 3.0–10.0, $T = 25^\circ\text{C}$ , $t = 2 \text{ h}$	Pseudo-II-order	Freundlich	Chemisorption	–	Hua et al. (2015)
Cd (II)	$q_{\max} = 13.02 \text{ mg g}^{-1}$ $q_{\max} = 5.3 \text{ mg g}^{-1}$ $q_{\max} = 9.2 \text{ mg g}^{-1}$ $q_{\max} = 0.043 \text{ mmol g}^{-1}$	$m = 10 \text{ g}$ , $C_0 = 1 \text{ mg L}^{-1}$ , pH = 3.0–10.0, $T = 25^\circ\text{C}$ , $t = 2 \text{ h}$ $m = 10 \text{ mg}$ , $C_0 = 10\text{--}200 \text{ mg L}^{-1}$ , pH = 6.0, $t = 24 \text{ h}$ $m = 0.1 \text{ g}$ , $C_0 = 0.04\text{--}0.8 \text{ mM}$ , pH = 7.0, $T = 25^\circ\text{C}$ , $t = 24 \text{ h}$	– Pseudo-II-order	Langmuir	Chemisorption Ion exchange	– –	Hua et al. (2015) Siswoyo et al. (2014)
Zn (II)	$q_{\max} = 46.000 \text{ mg kg}^{-1}$ $q_{\max} = 0.050 \text{ mmol g}^{-1}$	$m = 0.02\text{--}0.10 \text{ g}$ , $C_0 = 10\text{--}500 \text{ mg L}^{-1}$ , pH = 5.0–9.0, $t = 24 \text{ h}$ $m = 0.1 \text{ g}$ , $C_0 = 0.04\text{--}0.8 \text{ mM}$ , pH = 7.0, $T = 25^\circ\text{C}$ , $t = 24 \text{ h}$	Power function model Pseudo-I-order	Langmuir	Interaction between Cd(II) and –OH, O–Al–O entites Chemisorption	– –	Elkhatib et al. (2016) Silveti et al. (2015)
Pb (II)	$q_{\max} = 0.30 \text{ mmol g}^{-1}$ $q_{\max} = 0.26 \text{ mmol g}^{-1}$	$m = 10 \text{ g}$ , $C_0 = 0.4\text{--}8 \text{ mM}$ , pH = 2.0–9.0, $T = 25^\circ\text{C}$ , $t = 2 \text{ h}$ $m = 0.1 \text{ g}$ , $C_0 = 0.4\text{--}8 \text{ mM}$ , pH = 4.5, $T = 25^\circ\text{C}$ , $t = 24 \text{ h}$	– Pseudo-II-order	Freundlich Langmuir	Chemisorption	Endothermic	Zhou and Haynes (2010)
Cr (III)	$q_{\max} = 0.072 \text{ mmol g}^{-1}$ $q_{\max} = 0.37 \text{ mmol g}^{-1}$ $q_{\max} = 0.51 \text{ mmol g}^{-1}$	$m = 10 \text{ g}$ , $C_0 = 0.4\text{--}8 \text{ mM}$ , pH = 2.0–9.0, $T = 25^\circ\text{C}$ , $t = 2 \text{ h}$ $m = 10 \text{ g}$ , $C_0 = 0.4\text{--}8 \text{ mM}$ , pH = 2.0–9.0, $T = 25^\circ\text{C}$ , $t = 2 \text{ h}$	– Pseudo-II-order	Freundlich Langmuir	Chemisorption	– Endothermic	Castaldi et al. (2015) Zhou and Haynes (2010)
				Freundlich Langmuir	Chemisorption	– Endothermic	Zhou and Haynes (2011) Zhou and Haynes (2011)



**Table 2** (continued)

Adsorbate	Adsorption capacity ( $q_{max}$ )	Conditions for adsorption process	Kinetic model	Isotherm	Adsorption mechanism	Thermo-dynamic	References
Cr (VI)	$q_{max} = 0.21 \text{ mmol g}^{-1}$	$m = 10 \text{ g}$ , $C_0 = 0.4\text{--}8 \text{ mM}$ , $\text{pH} = 2.0\text{--}9.0$ , $T = 25 \text{ }^\circ\text{C}$ , $t = 2 \text{ h}$	Pseudo-II-order	Freundlich Langmuir	Chemisorption	Endothermic	Zhou and Haynes (2010)
	$q_{max} = 0.22 \text{ mmol g}^{-1}$	$m = 10 \text{ g}$ , $C_0 = 0.4\text{--}8 \text{ mM}$ , $\text{pH} = 2.0\text{--}9.0$ , $T = 25 \text{ }^\circ\text{C}$ , $t = 2 \text{ h}$	Pseudo-II-order	Freundlich Langmuir	Chemisorption	Endothermic	Zhou and Haynes (2011)
Hg (II)	$q_{max} = 75.4 \text{ mg g}^{-1}$	$C_0 = 80 \text{ mg L}^{-1}$ , $\text{pH} = 3.0\text{--}8.0$ , $T = 25 \text{ }^\circ\text{C}$ , $t = 96 \text{ h}$	Pseudo-I-order	Langmuir	Diffusion	–	Hovsepian and Bonzongo (2009)
Cu (II)	$q_{max} = 0.056 \text{ mmol g}^{-1}$	$m = 0.1 \text{ g}$ , $C_0 = 0.4\text{--}8 \text{ mM}$ , $\text{pH} = 4.5$ , $T = 25 \text{ }^\circ\text{C}$ , $t = 24 \text{ h}$	–	Freundlich	Chemisorption	–	Castaldi et al. (2015)
Co (II)	$q_{max} = 17.307 \text{ mg g}^{-1}$	$m = 0.5 \text{ g}$ , $C_0 = 0\text{--}800 \text{ mg L}^{-1}$ , $\text{pH} = 3.0\text{--}8.0$ , $T = 25 \text{ }^\circ\text{C}$ , $t = 48 \text{ h}$	Pseudo-II-order	Langmuir	Chemisorption	Endothermic	Jiao et al. (2017)
Methylene blue	$q_{max} = 70.4 \text{ mg g}^{-1}$	$m = 0.05\text{--}0.25 \text{ g}$ , $C_0 = 50\text{--}100 \text{ mg L}^{-1}$ , $\text{pH} = 3.0\text{--}9.0$ , $T = 20\text{--}25 \text{ }^\circ\text{C}$ , $t = 20\text{--}90 \text{ min}$	Pseudo-II-order	Langmuir	Electrostatic attraction	Exothermic	Rashed et al. (2016)

Jeon et al. (2018) concluded that adsorption mechanisms on raw AS were not only influenced by chemisorption, but also controlled by film diffusion due to natural organic material. Based on thermodynamic studies, they found that the adsorption of As on the AS is endothermic, i.e., the adsorption of As(V) increased with an increase in temperature and spontaneous, which was reflected from a negative value of the Gibbs free energy and a positive value of the standard enthalpy change. Also Kim et al. (2012) noticed that the adsorption of As can be controlled by pore diffusion and surface diffusion, because the AS has a high surface area ( $42.44 \text{ m}^2 \text{ g}^{-1}$ ) and narrow pore diameter (average 11.12 nm). Kim et al. (2012) and Nagar et al. (2010) also showed that the adsorption of As(V) onto the AS decreased as the pH of the solution increased. The highest adsorption capacity of AS for As(V) was observed at pH 3.8 (Kim et al. 2012).

High efficiency of AS for adsorption of As(III) and As(V) Caporale et al. (2013) also correlated with its high surface area ( $312.8\text{--}435.5 \text{ m}^2 \text{ g}^{-1}$ ). The AS they studied had a high organic matter content, and As(III) and As(V) were bound by humic substances. Additionally, Caporale et al. (2013) proposed several binding mechanisms, including the formation of ternary complexes with polyvalent cations (Al) forming a bridge between As anions and OM, the formation of outer sphere complexes with protonated amino groups of natural organic matter (NOM) and the formation of covalent bonds between phenolate/carboxylate groups of NOM and As(III) or As(V).

The adsorption capacity of AS for As(V) and As(III) was also determined by Makris et al. (2007). They used AS enriched with a small amount of a copolymer of sodium acrylate and acrylamide. After 48 h of reaction, the researchers obtained high removal efficiencies of both As(V) and As(III) at 93% and 55, respectively. A pore diffusion model was successfully applied by Makris et al. (2007) to explain As(III) and As(V) adsorption by activated alumina grains, while tested AS exhibited a Freundlich type As(III) and As(V) adsorption at concentrations up to  $3000 \text{ mg As L}^{-1}$ , implying huge affinity of the sludge surfaces for both As species. The researchers emphasized at the same time that AS is highly effective adsorption materials for As, and what is more the major advantage of using this material is that As(III) removal is not followed by a concomitant increase of As(V) in solution.

Similar conclusions were supported by the studies of the adsorption of As and Se on AS and desorption and regeneration, conducted by Zhou and Haynes (2012). Researchers hypothesized that As(V), As(III), Se(VI) and Se(IV) are

very strongly held to sludge surfaces, suggesting that they will not be readily leached from the material in a landfill, at least in the short term; moreover, they noticed that an alkaline environment is least favorable for adsorption of metal anions and favors desorption. In addition to the high pH being unfavorable for anion adsorption, the very high pH will favor dissolution of Al hydroxide surfaces thus releasing adsorbed anion from the surfaces. Additionally, they noted the decline in adsorption as pH was increased, suggesting that electrostatic repulsion was the dominant mechanism operative. Zhou and Haynes (2012) noticed that for all four oxyanions, their adsorption onto AS was rapid and 90%, or greater, of maximum adsorption had occurred after 120 min. The researchers suggested that chemisorption rather than diffusion/ion exchange was the rate-limiting step to adsorption; moreover, they showed that arsenate, arsenite and selenite have been form strong inner sphere complexes (bidentate and sometimes monodentate), while selenate forms only outer sphere complexes with Al oxide surface.

Ippolito et al. (2009) conducted studies on Se adsorption on AS, and using X-ray absorption spectroscopy they showed that Se(VI) and Se(IV) adsorption occurred as outer-sphere and inner-sphere complexes, respectively. They suggested that AS could be utilized to reduce Se concentrations in water regardless of redox conditions. And what is more, they hypothesized that AS appears to form stable complexes with adsorbed Se(IV) and Se(0) species, lessening the likelihood of the soluble Se(VI) release into the environment following re-oxidation of reduced Se species.

Lian et al. (2020) found that there is no report on the improvement in Mo(VI) adsorption efficiency by AS. Therefore, they decided to fill this gap by presenting the results of their research, in which they used thermally and acid activated AS for the adsorption of Mo(VI). The optimal condition of the activation the researchers ascertained as thermal activation at  $600 \text{ }^\circ\text{C}$  for 4 h followed by activation of hydrochloric acid at  $4.0 \text{ mol L}^{-1}$ . According to them, the maximum capacity of Mo(VI) adsorption increased from  $18.44 \text{ mg g}^{-1}$  before modification to  $39.52 \text{ mg g}^{-1}$  after modification. They showed that thermodynamic parameters indicated that the process of Mo(VI) adsorption was endothermic, entropy increasing and spontaneous. As the main mechanisms of Mo(VI) binding, Lian et al. (2020) have pointed to electrostatic interaction and ion exchange. The authors suggest that the modified AS is a green and recyclable adsorbent and could be used for the removal of Mo(VI) from wastewater, while encouraging more research in this direction.

Results of Hua et al. (2015) showed that AS is very effective low-cost adsorbents for oxyanions such as arsenate, vanadate and molybdate. They indicated that AS bound V, from single ion solution, in the highest amounts ( $q = 268.13 \text{ mmol kg}^{-1}$ ), then As ( $q = 247.31 \text{ mmol kg}^{-1}$ ) and in the smallest amounts—Mo ( $q = 94.65 \text{ mmol kg}^{-1}$ ). In their study, the maximum adsorption of Mo occurred at low pH with a sharp decline occurring between pH 4.0 and 6.0, while for V and As, adsorption remained at a maximum until around pH 8.0–10.0, followed by a sharp decline. They explained this with the fact that with increasing pH, the surface charge of AS becomes increasing negative resulting in greater electrostatic repulsion between anions and the adsorbent surface, and moreover, the adsorbent surfaces have a net negative charge above the point of zero charge, and at higher pH values, oxyanion adsorption may be inhibited due to increasing competitive effects of  $\text{OH}^-$  for adsorption surfaces. Their later research (Hua et al. 2018) on competitive adsorption of As, V and Mo onto AS was conducted using equimolar oxyanion concentrations in single, binary and tertiary combinations in adsorption isotherm and pH studies. They studied the desorption of previously adsorbed oxyanions in solutions containing single and double combinations of oxyanions. They observed that the presence of competing ions had a measurable effect in suppressing adsorption of Mo, V and As. Adsorption of Mo was decreased more by the presence of As than V, adsorption of As was suppressed a little by the presence of Mo, while adsorption of V was decreased a little by the presence of Mo. The authors observed a notably reduced adsorption of V on AS in the presence of As in the pH range 4–8, explaining the strong competition between these two oxyanions. Their studies have showed that desorption of Mo, V and As was greatest in the first desorption cycle and decreased with each successive cycle. Moreover, they observed that the order of selectivity in inducing desorption of the other oxyanions was  $\text{As} > \text{V} > \text{Mo}$  which is the same as the order of selectivity found in the adsorption and pH envelope experiments. The authors suggest that at equimolar concentrations of anions, in multielement solutions, As will be adsorbed preferentially and that it will tend to displace previously adsorbed V and more particularly Mo from adsorption sites.

**Cd and Zn adsorption onto AS** Among the trace metals, Cd(II) and Zn(II) are common industrial contaminants which simultaneously occur in most contaminated soils, which determines many problems related to metal contamination. Cd is considered an extremely toxic heavy metal and the main form of Cd in polluted water is Cd(II) (Elkhatib

et al. 2016; Genc-Fuhrman et al. 2007). In turn, Zn is relatively non-toxic; however, elevated Zn levels are harmful to the environment (Nachtegaal and Sparks 2004).

The potential of using AS as adsorbents for removing Cd from aqueous solution was investigated by Elkhatib et al. (2016), Silveti et al. (2015) and Siswoyo et al. (2014). Siswoyo et al. (2014) observed that the adsorption of the Cd ions on the AS depends on the nature of the adsorbent surface, and the distribution of metal species also depends on the pH of the solution. They observed that as the pH increased, there was an increase in metal ion removal, due to a decrease of  $\text{H}^+$  on the surface, which results in less metal ion repulsion, and what is more, the amounts of adsorbed Cd(II) increased with an increase in the adsorbent dosage due to the greater accessibility of the surface area or binding sites. Siswoyo et al. (2014) showed that the ideal pH for adsorption of  $10 \text{ mg L}^{-1}$  of Cd(II) ions in solution was within the range of 6. At the pH 6.0 and contact time 120 min, the maximum adsorption capacities were achieved. The researchers indicated the ion exchange model as a possible mechanism for Cd adsorption by the AS.

Elkhatib et al. (2016) used nanoparticles of AS as adsorbents to remove Cd(II) ions. They suggested that nanoparticles, compared to larger particles, are a more promising candidate for contaminated water and wastewater remediation. Their nanoparticles were prepared by using high-energy ball milling. The results of Cd(II) ions adsorption on three sizes of AS (2 mm, less than 51 microns and less than 100 nm) showed that the Cd<sup>2+</sup> adsorption efficiency increases with increasing adsorbent. Elkhatib et al. (2016) explain the increased adsorption with increasing adsorbent dose by the larger surface area and more adsorption sites available at the higher adsorbent dose. Moreover, the researchers observed that Cd(II) removal by tested adsorbent decreased as the pH increased from 5 to 9 and a maximum value was reached at an equilibrium pH of around 5. According to Elkhatib et al. (2016), the Cd(II) adsorption mechanism seems to be related to the interaction between Cd(II) and  $-\text{OH}$ ,  $\text{O}-\text{Al}-\text{O}$  surface groups.

However, Silveti et al. (2015) observed that the removal of Cd(II) and Zn(II) ions increased with increasing pH from 4.5 to 7.0. In their study, the maximum amount of Cd(II) and Zn(II) adsorbed by AS at pH 7.0 was more than twice than that recorded at pH 4.5. They observed that as the pH decreases, the AS surfaces become predominantly positively charged thus negatively influencing the adsorption of Cd(II) which is affected by Coulombic repulsion. Silveti et al. (2015) attribute the high Cd(II) and Zn(II) ions binding efficiency of the AS to the high content of the organic



and inorganic substances within AS which can act as coating, i.e., clogging the mesopores and micropores of Al (oxy) hydroxides. They further suggest that different types of association between surface Al nuclei and organic matter within AS cannot be excluded.

Suitability of AS for removal of heavy metals, including Zn(II), from an electroplating wastewater which had high concentrations of Cu and Cr along with other heavy metals was assessed by Ghorpade and Mansoor Ahammed (2018). Based on the batch adsorption study, the researchers noted that Zn(II) removal increased with increasing initial pH, while column test with real electroplating wastewater showed complete removal of zinc up to 100 bed volumes. The authors indicated that AS has the potential to be used as a filtration/adsorption medium for removal of metals from metal-bearing wastewaters.

**Pb, Cr, Cu, Co and Hg adsorption onto AS** Industrial wastewater is often contaminated with common heavy metals, such as Pb(II), Cr(III), Cr(VI), Cu(II), Co(II) and Hg(II) (Ghorpade and Mansoor Ahammed 2018). Of these metals, Cr, Cu, Pb and Hg are listed on US-EPA Priority Pollutants, with known negative impacts on both human health and ecological functions (US-EPA 2014).

AS from the water treatment plant in India, where polyaluminum chloride is used as a coagulant, has been used to remove Pb(II), Cr(VI), Cu(II), Co(II) and Hg(II) from aqueous solutions (Ghorpade and Mansoor Ahammed 2018). The authors noted that for all metals tested there was initially rapid adsorption and slowed down to a constant value, while for Pb, almost complete sorption occurred within 15 min. They reported that Pb(II) could form strong inner sphere complexes with aluminum oxide surfaces, what could be responsible for complete adsorption of Pb(II). Moreover, they observed that Cu(II), Co(II) and Hg(II) removal increased with increase in initial pH, whereas for Pb(II) there was no influence of pH and almost complete removal occurred at all the pH values tested, while Cr(VI) presented a different trend with higher removal at lower pH values of 2.5–4.0 and then decreased with increase in pH, which is derived from the  $\text{pH}_{\text{PZC}}$  value of the adsorbent under study. In addition, they noted the effect of adsorbent dose on the amounts of heavy metals removed. Ghorpade and Mansoor Ahammed (2018) concluded that the AS they studied showed high binding capacities of the heavy metals studied.

Moreover, according to Zhou and Haynes (2010) studies adsorption of Pb(II), Cr(III) and Cr(VI) increased with increasing equilibrium concentration of the metal ion and reached a maximum value as the adsorbent became

progressively saturated. The authors found that the adsorption process was rapid and at a contact time of 120 min, 90% or greater of maximum adsorption had occurred, and the pseudo-second-order equation gave a better fit to the experimental kinetic data, what suggested that chemisorption rather than diffusion/ion exchange was the rate-limiting step to adsorption. In addition, the researchers found that adsorption edges for  $\text{Cr}^{3+}$  and  $\text{Pb}^{2+}$  showed increasing adsorption from near zero to near complete over a relatively small pH range. They explained that the variable charge on hydroxyl-Al surfaces arises from protonation and deprotonation of the potential determining  $\text{M-OH}_2^{0.5+}$  and  $\text{M-OH}^{0.5-}$  groups; the surface becomes increasingly negatively charged as hydroxyl ion activity (and pH) increases, thus favoring specific adsorption of cations. In addition, they observed that, as pH is raised, hydrolysis of hydrated cations proceeds and their adsorption generally occur preferentially as hydrolyzed ions. By contrast, they noticed that Cr(VI) adsorption showed a maximum between pH 3 and 4 and declined thereafter with increasing pH.

These researchers (Zhou and Haynes 2011) in subsequent years compared the adsorption properties of two different sediments from two separate water treatment stations in relation to Cr(III), Cr(VI) and Pb(II). The sludges they tested differed in their physicochemical properties, and the biggest differences were noted in the magnitude of the specific surface area, which was  $97.3 \text{ m}^2 \text{ g}^{-1}$  and  $290.1 \text{ m}^2 \text{ g}^{-1}$ , respectively, due to the presence of activated C in one of the tested materials. Based on this study, the researchers again noted that adsorption appeared to be strongly pH-dependent. Zhou and Haynes (2011) proved again that AS is a suitable material for developing a low-cost adsorbent for removing Pb(II), Cr(III) and Cr(VI) from aqueous solutions. Also the column studies by Ghorpade and Ahammed (2018) showed the high efficiency of AS for removing Cr(VI).

AS as an adsorbent for removing Co(II) and Pb(II) from aqueous solution was the subject of a study by Castaldi et al. (2015). Their kinetic tests showed that Pb(II) adsorption by the AS was biphasic, highlighting an initially fast adsorption, followed by a slower Pb(II) sorption rate. They observed that after the first hour, more than 85% of Pb(II) was adsorbed by AS, while the amount of adsorbed Cu(II) accounted for about 60% of the total amount of adsorbed copper after 24 h. The authors obtained higher adsorption of Pb(II) by tested AS compared to Cu(II). In addition, based on the sequential chemical extraction performed, the researchers concluded that the specific adsorption of both cations seems to play a more important role than nonspecific adsorption; in particular, the Cu(II), even if less adsorbed, was more strongly



retained than Pb(II) by the colloidal component of AS. The authors suggested that the mechanism which regulated the adsorption of Cu(II) and Pb(II) by the AS could be the result of chemical interactions that gave rise to the formation of inner-sphere surface complexes. Additionally, Castaldi et al. (2015) explain a strong contribution to this adsorption mechanism is derived from the organic matter incorporated in the tested material.

Hovsepyan and Bonzongo (2009) used AS to efficiently adsorb and immobilize Hg from aqueous solutions. The tested material with average specific surface area of  $48 \text{ m}^2 \text{ g}^{-1}$  and internal micropore surface area of  $120 \text{ m}^2 \text{ g}^{-1}$  is used in a series of batch adsorption experiments, and they noticed that the obtained adsorption isotherms indicated a strong affinity of Hg for tested material. The authors observed that the adsorption isotherms showed a strong affinity of Hg for AS. Moreover, the researchers noted that the adsorption kinetic data were best fit to a pseudo-first-order model, while the use of the Weber–Morris and Bangham models suggested that the intraparticle diffusion could be the rate-limiting step for Hg immobilization. However, the authors clearly indicated the need for further research for more in depth evaluation of Hg adsorption mechanisms and the long-term stability of formed Hg–[AS] complexes and at the same time suggested that the results from these short-term experiments demonstrate that tested material can be effectively used to remove Hg from aqueous solutions. The high need to expand research on the mechanisms of adsorption of metals, including Hg, by AS was reported by Quinones et al. (2016) in their manuscript, and they made it clear that knowledge of the mechanisms of metal sorption by AS remains very limited and there is a lack of data on the long-term stability of the resulting metal-AS complexes as a function of changing key environmental parameters.

Jiao et al. (2017) observed that the adsorption of Co(II) on AS was dependent on pH values and was affected by the ionic strength. In addition, they showed that Co(II) adsorption was a spontaneous endothermic process and was favorable at high temperature. They observed that the desorption of Co(II) depended on the initial solution pH, and only minimal amounts of adsorbed Co(II) were desorbed. Based on the FT-IR spectra performed, Jiao et al. (2017) indicate that Co(II) interacted with the AS surface through strong covalent bonding with Fe(Al)-O functional groups.

### Adsorption of dyes using AS

One of the most serious problems of the water environment in modern society is water pollution by synthetic dyes (Gadekar and Mansoor Ahammed 2020; Nourmoradi et al. 2016; Yusuff et al. 2017). These substances are widely used

for dyeing and printing in various industries. Huge amounts of colored wastewater are discharged by various sectors such as textiles, leather, printing, laundry, tanning, rubber, plastic, painting, etc. (Geng et al. 2018; Rashed et al. 2016). Over 100,000 commercially available dyes exist, and more than  $7 \times 10^5$  tonnes per year is produced annually, and what is more 5–10% of the dye stuff is lost in the industrial effluents (Butani and Mane 2017; Gadekar and Mansoor Ahammed 2016; Rashed et al. 2016). Due to their chemical structure, dyes belong to the groups such as azo, anthraquinone, oxazine, acridine, formazan, phthalocyanine, thiazine, triphenylmethane, triarylmethane, nitroso and nitro, etc. and due to their application to reactive, acidic, cationic, direct, vat, disperse, solvent and sulfur (Pajak 2021). The high solubility of dyes in water causes their wide spread in the environment; moreover, they are often durable, resistant to degradation, which makes them harmful to crops, aquatic life and, importantly, they are toxic, carcinogenic and mutagenic for a humans (Chu 2001; Md Nor et al. 2014; Pajak 2021; Poormand et al. 2017; Zhao et al. 2011). That is why, it is so important to effectively remove dyes from wastewater before it is introduced into the sewage system and the environment. As shown in the available literature, dyes can be successfully removed using alum sludge.

Md Nor et al. (2014) conducted research on the kinetics of the Remazol blue R dye (cationic dye) adsorption by dewatered AS from a local water treatment plant located in Putrajaya, Malaysia. They conducted kinetics studies in a batch system, mixing 40 g of AS in 200 mL of  $100 \text{ mg L}^{-1}$  synthetic wastewater, and then applied Elovich kinetic models, pseudo-second-order type I, II and intramolecular diffusion. Their results showed that the adsorption kinetic dye was best described by a pseudo-second-order type I kinetic model with  $R^2$  of 0.994. According to Md Nor et al. (2014), the adsorption rate was controlled by a chemical adsorption.

The best known and most frequently used dye for dyeing cotton, silk, wool, Chi-Liang, paper and as a coating for papers is Methylene Blue. As many researchers note (Liu et al. 2016; Rashed et al. 2016; Poormand et al. 2017), Methylene Blue can cause some harmful effects such as a cute exposure to shock, Heinz body formation, cyanosis, jaundice, quadriplegia and tissue necrosis in humans. Therefore, steps and preventive measures should be taken to prevent this dye from leaking into the environment. For this reason, Yusuff et al. (2017) conducted a study on the possibility of adsorption of this dye on AS. The researchers prepared the adsorbent from a fresh drinking water treatment sludge cake which was obtained from a local waste water treatment plant. The tested AS was heat treated at high temperature (100, 450 and 800 °C). During the study on the adsorption



capacity of AS, they took into account 6 parameters of the adsorption process, i.e., the adsorbent treatment temperature, the adsorbent dose, the initial concentration of the dye in the solution, the pH of the dye solution, the contact time of the solid phase with the solution and the temperature during the adsorption process. In addition, they subjected the experimental data to second-order polynomial regression analysis to analyze the experimental data by estimating the response as a function of the independent variables. On the basis of the calculated parameters, Yusuff et al. (2017) observed that the most favorable conditions, when the removal efficiency of Methyl Blue by AS was 100%, are under the following conditions: the temperature of treatment, pH of solution, sorbent dosage, initial dye concentration, contact period and temperature of sorption were 450 °C, 6.0, 1.55 g, 150 mg L<sup>-1</sup>, 120 min and 52.5 °C, respectively.

Rashed et al. (2016) used the AS from the Fatera drinking water treatment plant in Kom Ombo city, Aswan governorate, Egypt, in adsorption of the Methylene Blue dye. The study conducted adsorption experiments under different conditions of initial dye concentration (50–100 mg L<sup>-1</sup>), adsorbent dosage (0.05–0.25 g), solution pH (3.0–9.0), temperature (20–60 °C) and contact time (20–90 min). The authors suggested that the low adsorption of Methylene Blue at an acidic pH was due to the presence of excess H<sup>+</sup> ions that compete with the dye cation for adsorption sites. Rashed et al. (2016) noted that the number of positively charged sites decreased, while the number of negatively charged sites increased, which favored the adsorption of Methylene Blue due to electrostatic attraction. Moreover, they observed that as the pH value increased from 7.0 to 9.0, the efficiency of the dye removal decreased, and at higher solution pH, the decrease in the adsorption rate was due to the formation of a hydroxyl complex between the adsorbent and the dye. Moreover, they observed the decrease in adsorption with the rise in temperature, what may have resulted from the weakening of the adsorptive forces between the active sites of the adsorbents and the adsorbate species. Their results indicated also that the Langmuir isotherm is most appropriate for the adsorption, indicating that the monolayer of dye molecules covers the adsorbent sludge surface. In turn, on the basis of kinetic parameters and the coefficient of determination, they noticed that the adsorption process proceeded in accordance with the pseudo-second-order equation.

AS has also proved to be an effective adsorbent for removing textile dyes from real textile wastewater. Such research was undertaken by Gadekar and Mansoor Ahammed (2020). Researchers used as an adsorbent in their work AS, which were collected from the coagulation/flocculation unit of a water treatment plant in Bhandup, Mumbai, India, where

polyaluminum chloride (PACl) is used as a coagulant, whereas textile dye wastewater was collected from a common effluent treatment plant run by Palsana Enviro Care, Surat, Gujarat, India. The authors explained that this plant treats wastewater collected from different textile dye industries, where disperse dyes are mostly use in their processes. They conducted batch adsorption studies using different adsorbent doses (in the range of 10–50 g L<sup>-1</sup>) with 50 mL of wastewater, the pH of the wastewater in the range of 3.0–7.0, and the contact time of the solid phase with the solution at 60 min. Their results showed a maximum color removal in batch adsorption and column operation of 36% and 60%, respectively. The researchers obtained maximum color removal at an initial pH of 3.0, while in column tests, the total adsorbent capacity of the adsorbent is obtained at 180 and 120 bed volumes for an initial pH of 3.0 and 6.2, respectively. As noted by Gadekar and Mansoor Ahammed (2020), though the capacity of the AS would be less compared to commonly available adsorbents, its availability in large quantities at free of cost can be used advantageously. Gadekar and Mansoor Ahammed (2019) in their previous work showed that AS is an effective adsorbent for removing disperse dye from aqueous solutions. Under optimal conditions, i.e., pH 3.0, AS dose of 30 g L<sup>-1</sup> and dye concentration of 75 mg L<sup>-1</sup>, they achieved color removal of 52.6 ± 2.0%. The researchers indicated that adsorption onto AS could be used as a primary treatment for removal of color from dye wastewater.

## Conclusion

In this review, a large amount of literature has been collected, on the basis of which the adsorption properties, binding mechanism, adsorption isotherms, etc. of Al-based post-coagulation sludge in relation to inorganic pollutants, i.e., As, Se, V, Mo, Cd, Zn, Pb, Cr, Cu, Co, Hg, and organic ones, i.e., textile dyes. Moreover, the formation and characterization of AS, as well as its physicochemical properties, are presented.

On the basis of the collected literature, it was observed that the amount, composition and properties of AS strongly depend on the quality and quantity of the treated water, the type and dose of coagulants used (aluminum salts used in the treatment of surface waters and iron salts used in the treatment of groundwater) and the process conditions; therefore, depending on the above factors, the physicochemical properties of the AS differ from each other. It was noted that AS is characterized by a large specific surface which, according to the researchers, may range from 21 to 364.55 m<sup>2</sup> g<sup>-1</sup>, which



is about 1/3 of the surface of activated carbon, high porosity, irregularity and a loose structure with limited strength, thanks to which this material has a high adsorption capacity to both inorganic and organic pollutants.

Analyzing the collected results of the adsorption process, it was observed that in most cases the Freundlich and Langmuir isotherms described the adsorption of the inorganic pollutants well and the Langmuir isotherm of organic pollutants. Moreover, it was noted that the adsorption process most often followed a pseudo-second-order model. The researchers most often pointed to chemisorption as the mechanism of metal ions adsorption on AS and electrostatic interactions of dyes. On the basis of the presented results of thermodynamic studies, it was noted that the adsorption of metal ions on AS is endothermic, while the adsorption of dyes is exothermic.

On the basis of the collected literature, it can be concluded that the tested adsorbent material is an effective and economical adsorbent for removing textile dyes and inorganic pollutants from water and wastewater, both in the study of single pollutants and many pollutants.

**Acknowledgements** The study was conducted in the Institute of Environmental Engineering of the Polish Academy of Sciences

## Declarations

**Conflict of interest** The author declare that have no conflict of interest.

**Ethical approval** This article does not contain any studies with human participants or animals performed by any of the authors.

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## References

- Ahmad T, Ahmad K, Alam M (2016) Characterization of water treatment plant's sludge and its safe disposal options. *Procedia Environ Sci* 35:950–955. <https://doi.org/10.1016/j.proenv.2016.07.088>
- Awab H, Paramalinggam PTP, Yusoff ARM (2012) Characterization of alum sludge for reuse and disposal. *MJFAS* 8:251–255. <https://doi.org/10.11113/mjfas.v8n4.160>
- Babatunde AO, Zhao YQ (2007) Constructive approaches towards water treatment works sludge management: an international review of beneficial re-uses. *Crit Rev Environ Sci Technol* 37:129–164. <https://doi.org/10.1080/10643380600776239>
- Babatunde AO, Zhao YQ (2010) Equilibrium and kinetic analysis of phosphorus adsorption from aqueous solution using waste alum sludge. *J Hazard Mater* 184:746–752. <https://doi.org/10.1016/j.jhazmat.2010.08.102>
- Babatunde AO, Zhao YQ, Yang Y, Kearney P (2008) Reuse of dewatered aluminium-coagulated water treatment residual to immobilize phosphorus: batch and column trials using a condensed phosphate. *Chem Eng J* 136:108–115. <https://doi.org/10.1016/j.cej.2007.03.013>
- Babatunde AO, Zhao YQ, Burke AM, Morris MA, Hanrahan JP (2009) Characterization of aluminium-based water treatment residual for potential phosphorus removal in engineered wetlands. *Environ Pollut* 157:2830–2836. <https://doi.org/10.1016/j.envpol.2009.04.016>
- Bai L, Wang Ch, He L, Pei Y (2014) Influence of the inherent properties of drinking water treatment residuals on their phosphorus adsorption capacities. *J Environ Sci* 26:2397–2405. <https://doi.org/10.1016/j.jes.2014.04.002>
- Balkaya M (2015) Evaluation of the use of alum sludge as hydraulic barrier layer and daily cover material in landfills: a finite element analysis study. *Desalin Water Treat* 57:1–13. <https://doi.org/10.1080/19443994.2015.1005154>
- Barakwan RA, Trihadiningrum Y, Bagastyo AY (2019) Characterization of alum sludge from Surabaya Water Treatment Plant, Indonesia. *J Ecol Eng* 20:7–13. <https://doi.org/10.12911/22998993/104619>
- Boretti A, Rosa L (2019) Reassessing the projections of the World Water Development Report. *Npj Clean Water* 2:15. <https://doi.org/10.1038/s41545-019-0039-9>
- Butani SA, Mane SJ (2017) Coagulation/flocculation process for cationic and anionic dye removal using water treatment residuals. *IJES* 6:1–5
- Caporale AG, Punamiya P, Pigna M, Violante A, Sarkar D (2013) Effect of particle size of drinking-water treatment residuals on the sorption of arsenic in the presence of competing ions. *J Hazard Mater* 260:644–651. <https://doi.org/10.1016/j.jhazmat.2013.06.023>
- 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. <https://doi.org/10.1016/j.jhazmat.2013.10.037>
- Castaldi P, Silveti M, Garau G, Demurtas D, Deiana S (2015) Copper(II) and lead(II) removal from aqueous solution by water treatment residues. *J Hazard Mater* 283:140–147. <https://doi.org/10.1016/j.jhazmat.2014.09.019>
- Chiang YW, Ghyselbrecht K, Santos RM, Martens JA, Swennen R, Cappuyns V, Meesschaert B (2012) Adsorption of multi-heavy metals onto water treatment residuals: sorption capacities and applications. *Chem Eng J* 200:405–415. <https://doi.org/10.1016/j.cej.2012.06.070>
- Chi-Liang Y, Dyi-Hwa T, Tung-T L (2011) Characterization of eco-cement paste produced from sludge. *Chemosphere* 84:220–226. <https://doi.org/10.1016/j.chemosphere.2011.04.050>
- Chu W (2001) Dye removal from textile dye wastewater using recycled alum sludge. *Water Res* 35:3147–3152. [https://doi.org/10.1016/S0043-1354\(01\)00015-X](https://doi.org/10.1016/S0043-1354(01)00015-X)
- Crini G (2006) Non-conventional low-cost adsorbents for dye removal: a review. *Bioresour Technol* 60:67–75. <https://doi.org/10.1016/j.biortech.2005.05.001>
- 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 Manag* 38:321–335. <https://doi.org/10.1016/j.wasman.2014.11.025>
- Dayton EA, Basta NT (2005) A method for determining the phosphorus sorption capacity and amorphous aluminium of aluminium-based



- drinking water treatment residuals. *J Environ Qual* 34:1112–1118. <https://doi.org/10.2134/jeq2004.0230>
- Elkhatib E, Mahdy A, Sherif F, Hamadeen H (2015) Evaluation of a novel water treatment residual nanoparticles as a sorbent for arsenic removal. *J Nanomater*. <https://doi.org/10.1155/2015/912942>
- Elkhatib E, Mahdy A, Sherif F, Elshemy W (2016) Competitive adsorption of cadmium(II) from aqueous solutions onto nanoparticles of water treatment residual. *J Nanomater*. <https://doi.org/10.1155/2016/8496798>
- Gadekar MR, Mansoor Ahammed M (2016) Coagulation/flocculation process for dye removal using water treatment residuals: modelling through artificial neural networks. *Desalin Water Treat* 57:26392–26400. <https://doi.org/10.1080/19443994.2016.1165150>
- Gadekar MR, Mansoor Ahammed M (2019) Modelling dye removal by adsorption onto water treatment residuals using combined response surface methodology-artificial neural network approach. *J Environ Manag* 231:241–248. <https://doi.org/10.1016/j.jenvman.2018.10.017>
- Gadekar MR, Mansoor Ahammed M (2020) Use of water treatment residuals for colour removal from real textile dye wastewater. *Appl Water Sci* 10:160. <https://doi.org/10.1007/s13201-020-01245-9>
- Genc-Fuhrman H, Mikkelsen PS, Ledin A (2007) Simultaneous removal of As, Cd, Cr, Cu, Ni and Zn from stormwater: experimental comparison of 11 different sorbents. *Water Res* 41:591–602. <https://doi.org/10.1016/j.watres.2006.10.024>
- Geng Y, Zhang J, Zhou J, Lei J (2018) Study on adsorption of methylene blue by a novel composite material of TiO<sub>2</sub> and alum sludge. *RSC Adv* 8:32799–32807. <https://doi.org/10.1039/c8ra05946b>
- Georgantas DA, Grigoropoulou HP (2005) Phosphorus removal from synthetic and municipal wastewater using spent alum sludge. *Water Sci Technol* 52:525–532. <https://doi.org/10.2166/wst.2005.0732>
- Ghorpade A, Mansoor Ahammed M (2018) Water treatment sludge for removal of heavy metals from electroplating wastewater. *Environ Eng Res* 23:92–98. <https://doi.org/10.4491/eer.2017.065>
- Gibbons MK, Gagnon GA (2011) Understanding removal of phosphate or arsenate onto water treatment residual solids. *J Hazard Mater* 186:1916–1923. <https://doi.org/10.1016/j.jhazmat.2010.12.085>
- Guan X-H, Chen G-H, Shang C (2005) Re-use of water works sludge to enhance particulate pollutant removal from sewage. *Water Res* 39:3433–3440. <https://doi.org/10.1016/j.watres.2004.07.033>
- Gupta VK, Suhas (2009) Application of low-cost adsorbents for dye removal—a review. *J Environ Manag* 90:2313–2342. <https://doi.org/10.1016/j.jenvman.2008.11.017>
- Hargreaves AJ, Vale P, Whelan J, Alibardi L, Constantino C, Dotro G, Cartmell E, Campo P (2018) Impacts of coagulation-flocculation treatment on the size distribution and bioavailability of trace metals (Cu, Pb, Ni, Zn) in municipal wastewater. *Water Res* 128:120–128. <https://doi.org/10.1016/j.watres.2017.10.050>
- Hou QJ, Meng PP, Pei HY, Hu WR, Chen Y (2018) Phosphorus adsorption characteristics of alum sludge: adsorption capacity and the forms of phosphorus retained in alum sludge. *Matter Lett* 229:31–35. <https://doi.org/10.1016/j.matlet.2018.06.102>
- Hovsepyan A, Bonzongo J-CJ (2009) Aluminium drinking water treatment residuals (Al-WTRs) as sorbent for mercury: implications for soil remediation. *J Hazard Mater* 164:73–80. <https://doi.org/10.1016/j.jhazmat.2008.07.121>
- Hua T, Haynes RJ, Zhou Y-F, Boulemant A, Chandrawana I (2015) Potential for use of industrial waste materials as filter media for removal of Al, Mo, As, V and Ga from alkaline drainage in constructed wetlands—adsorption studies. *Water Res* 71:32–41. <https://doi.org/10.1016/j.watres.2014.12.036>
- Hua T, Haynes RJ, Zhou Y-F (2018) Competitive adsorption and desorption of arsenate, vanadate, and molybdate onto the low-cost adsorbent materials alum water treatment sludge and bauxite. *Environ Sci Pollut Res* 25:34053–34062. <https://doi.org/10.1007/s11356-018-3301-7>
- Ippolito JA, Barbarick KA, Heil DM, Chandler JP, Redente EF (2003) Phosphorus retention mechanisms of a water treatment residual. *J Environ Qual* 32:1857–1864. <https://doi.org/10.2134/jeq2003.1857>
- Ippolito JA, Scheckel KG, Barbarick KA (2009) Selenium adsorption to aluminium-based water treatment residuals. *J Colloid Interface Sci* 338:48–55. <https://doi.org/10.1016/j.jcis.2009.06.023>
- Ippolito JA, Barbarick KA, Elliott HA (2011) Drinking water treatment residuals: a review of recent uses. *J Environ Qual* 40:1–12. <https://doi.org/10.2134/jeq2010.0242>
- Jeon E-K, Ryu S, Park S-W, Wang L, Tsang DCW, Baek K (2018) Enhanced adsorption of arsenic onto alum sludge modified by calcination. *J Clean Prod* 176:54–62. <https://doi.org/10.1016/j.jclepro.2017.12.153>
- Jiao J, Zhao J, Pei Y (2017) Adsorption of Co(II) from aqueous solutions by water treatment residuals. *J Environ Sci* 52:232–239. <https://doi.org/10.1016/j.jes.2016.04.012>
- Jo JY, Choi JH, Tsang YF, Baek K (2021) Pelletized adsorbent of alum sludge and bentonite for removal of arsenic. *Environ Pollut* 277:116747. <https://doi.org/10.1016/j.envpol.2021.116747>
- Johnson OA, Napiah M, Kamaruddin I (2014) Potential uses of waste sludge in construction industry: a review. *Res J Appl Sci* 8:565–570. <https://doi.org/10.19026/rjaset.8.1006>
- Jordán MM, Almendro-Candel MB, Romero M, Rincón JM (2005) Application of sewage sludge in the manufacturing of ceramic tile bodies. *Appl Clay Sci* 30:219–224. <https://doi.org/10.1016/j.clay.2005.05.001>
- Keeley J, Jarvis P, Judd SJ (2014) Coagulant recovery from water treatment residuals: a review of applicable technologies. *Environ Sci Technol* 44:2675–2719. <https://doi.org/10.1080/10643389.2013.829766>
- Kim JG, Kim JH, Moon H, Chon C, Ahn JS (2002) Removal capacity of water plant alum sludge for phosphorus in aqueous solution. *Chem Speciat Bioavailab* 14:67–73. <https://doi.org/10.3184/095422902782775344>
- Kim Y-S, Kim D-H, Yang J-S, Baek K (2012) Adsorption characteristics of As(III) and As(V) on alum sludge from water purification facilities. *Sep Sci Technol* 47:2211–2217. <https://doi.org/10.1080/01496395.2012.700676>
- Kim JG, Kim HB, Yoon GS, Kim SH, Min SJ, Tsang DCW, Baek K (2020) Simultaneous oxidation and adsorption of arsenic by one-step fabrication of alum sludge and graphitic carbon nitride (g-C<sub>3</sub>N<sub>4</sub>). *J Hazard Mater* 383:121138. <https://doi.org/10.1016/j.jhazmat.2019.121138>
- Krishna KCB, Aryal A, Jansen T (2016) Comparative study of ground water treatment plants sludges to remove phosphorous from wastewater. *J Environ Manag* 180:17–23. <https://doi.org/10.1016/j.jenvman.2016.05.006>
- Kumar R, Kang C-U, Mohan D, Khan MA, Lee J-H, Lee SS, Jeon B-H (2020) Waste sludge derived adsorbents for arsenate removal from water. *Chemosphere* 239:124832. <https://doi.org/10.1016/j.chemosphere.2019.124832>
- Lee LY, Wang B, Guo H, Hu JY, Ong SL (2015) Aluminum-based water treatment residue reuse for phosphorus removal. *Water* 7:1480–1496. <https://doi.org/10.3390/w7041480>
- Lian J, Zhou F, Chen B, Yang M, Wang S, Liu Z, Niu S (2020) Enhanced adsorption of molybdenum(VI) onto drinking water treatment residues modified by thermal treatment and acid activation. *J Clean Prod* 244:118719. <https://doi.org/10.1016/j.jclepro.2019.118719>
- Liu R, Zhao Y, Sibille C, Ren B (2016) Evaluation of natural organic matter release from alum sludge reuse in wastewater treatment

- and its role in P adsorption. *Chem Eng J* 302:120–127. <https://doi.org/10.1016/j.cej.2016.05.019>
- Lombi E, Stevens DP, McLaughlin MJ (2010) Effect of water treatment residuals on soil phosphorus, copper and aluminum availability and toxicity. *Environ Pollut* 158:2110–2116. <https://doi.org/10.1016/j.envpol.2010.03.006>
- Makris KC, Harris WG, O'connor GA, Obreza TA, Elliott HA (2005) Physicochemical properties related to long-term phosphorus retention by drinking-water treatment residuals. *Environ Sci Technol* 39:4280–4289. <https://doi.org/10.1021/es0480769>
- Makris KC, Sarkar D, Datta R (2006) Aluminium-based drinking-water treatment residuals: a novel sorbent for perchlorate removal. *Environ Poll* 140:9–12. <https://doi.org/10.1016/j.envpol.2005.08.075>
- Makris KC, Sarkar D, Parsons JG, Datta R, Gardea-Torresdey JL (2007) Surface arsenic speciation of a drinking-water treatment residual using X-ray absorption spectroscopy. *J Colloid Interface Sci* 311:544–550. <https://doi.org/10.1016/j.jcis.2007.02.078>
- Maqbool N, Khan Z, Asghar A (2015) Reuse of alum sludge for phosphorus removal from municipal wastewater. *Desalin Water Treat* 57:13246–13254. <https://doi.org/10.1080/19443994.2015.1055806>
- Md Nor MA, Ong KK, Mohamad S, Ahmad Nasaruddin NA, Jamari NLA, Wan Yunus WMZ (2014) Kinetic study of a cationic dye adsorption by dewatered alum sludge. *Mater Res Innov* 18:S6-140-S6-143. <https://doi.org/10.1179/1432891714Z.00000000945>
- Nachtegaal M, Sparks DL (2004) Effect of iron oxide coatings on zinc sorption mechanisms at the clay-mineral/water interface. *J Colloid Interface Sci* 276:13–23. <https://doi.org/10.1016/j.jcis.2004.03.031>
- 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* 78:1028–1035. <https://doi.org/10.1016/j.chemosphere.2009.11.034>
- Nair AT, Mansoor Ahammed M (2015) The reuse of water treatment sludge as a coagulant for post-treatment of UASB reactor treating urban wastewater. *J Clean Prod* 96:272–281. <https://doi.org/10.1016/j.jclepro.2013.12.037>
- Nourmoradi H, Zabihollahi S, Pourzamani HR (2016) Removal of a common textile dye, navy blue (NB), from aqueous solutions by combined process of coagulation–flocculation followed by adsorption. *Desalin Water Treat* 57:5200–5211. <https://doi.org/10.1080/19443994.2014.1003102>
- 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. <https://doi.org/10.1016/j.jenvman.2010.11.016>
- Ong DC, Kan CC, Pingul-Ong SMB, de Luna MDG (2017) Utilization of groundwater treatment plant (GWTP) sludge for nickel removal from aqueous solutions: isotherm and kinetic studies. *J Environ Chem Eng* 5:5746–5753. <https://doi.org/10.1016/j.jece.2017.10.046>
- Pajak M (2021) Adsorption capacity of smectite clay and its thermal and chemical modification for two anionic dyes: comparative Study. *Water Air Soil Pollut* 232:83. <https://doi.org/10.1007/s11270-021-05032-3>
- Poormand H, Leili M, Khazaei M (2017) Adsorption of methylene blue from aqueous solutions using water treatment sludge modified with sodium alginate as a low cost adsorbent. *Water Sci Technol* 75:281–295. <https://doi.org/10.2166/wst.2016.510>
- Qi L, Cheng R, H-ch W, Zheng X, Zhang G-m, Li G-b (2011) Recycle of alum sludge with PAC (RASP) for drinking water treatment. *Desalin Water Treat* 25:170–175. <https://doi.org/10.5004/dwt.2011.1603>
- Quinones KD, Hovsepian A, Oppong-Anane A, Bonzongo JCJ (2016) Insights into the mechanisms of mercury sorption onto aluminum based drinking water treatment residuals. *J Hazard Mater* 307:184–192. <https://doi.org/10.1016/j.jhazmat.2016.01.001>
- Rashed MN, El Taher MAE-D, Fadlalla SMM (2016) Adsorption of methylene blue using modified adsorbents from drinking water treatment sludge. *Water Sci Technol* 74:1885–1898. <https://doi.org/10.2166/wst.2016.377>
- Razali M, Zhao Y, Bruen M (2007) Effectiveness of a drinking-water treatment sludge in removing different phosphorus species from aqueous solution. *Sep Purif Technol* 55:300–306. <https://doi.org/10.1016/j.seppur.2006.12.004>
- Ren B, Lyczko N, Zhao Y, Nzihou A (2021) Simultaneous hydrogen sulfide removal and wastewater purification in a novel alum sludge-based odor-gas aerated biofilter. *Chem Eng J* 419:129558. <https://doi.org/10.1016/j.cej.2021.129558>
- Scalize PS, Souza LMD, Albuquerque A (2019) Reuse of alum sludge for reducing flocculant addition in water treatment plants. *Environ Prot Eng* 45:57–70. <https://doi.org/10.5277/epe190105>
- 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* 226:313. <https://doi.org/10.1007/s11270-015-2578-0>
- Siswoyo E, Mihara Y, Tanaka S (2014) Determination of key components and adsorption capacity of a low cost adsorbent based on sludge of drinking water treatment plant to adsorb cadmium ion in water. *Appl Clay Sci* 97–98:146–152. <https://doi.org/10.1016/j.clay.2014.05.024>
- Soleha MY, Ong KK, Wan MdZin WY, Mansor A, Anwar F, Azowa IN, Shafiq SASM, Aisyah ASN, Aidy A, Ku Zarina KA, Teoh CC (2016) Characterization of raw and thermally treated alum sludge. *Key Eng Mater* 701:138–142. <https://doi.org/10.4028/www.scientific.net/KEM.701.138>
- Tarrago M, Garcia-Valles M, Aly MH, Martínez S (2017) Valorization of sludge from a wastewater treatment plant by glass-ceramic production. *Ceram Int* 43:930–937. <https://doi.org/10.1016/j.ceramint.2016.10.083>
- Tugrul AB, Hacıyakupoglu S, Erenturk SA, Karatepe N, Baytas AF, Altinsoy N, Baydogan N, Buyuk B, Demir E (2013) Selenium adsorption on activated carbon by using radiotracer technique. In: Dincer I, Colpan C, Kadioglu F (eds) Causes, impacts and solutions to global warming. Springer, New York
- 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 Poll* 230:227. <https://doi.org/10.1007/s11270-019-4272-0>
- US-EPA (2014) Priority Pollutant List. <https://www.epa.gov/sites/default/files/2015-09/documents/priority-pollutant-list-epa.pdf>
- Wang W, Ma C, Zhang Y, Yang S, Shao Y, Wang X (2016) Phosphate adsorption performance of a novel filter substrate made from drinking water treatment residuals. *J Environ Sci* 45:191–199. <https://doi.org/10.1016/j.jes.2016.01.010>
- Wołowicz M, Komorowska-Kaufman M, Pruss A, Rzepa G, Bajda T (2019) Removal of heavy metals and metalloids from water using drinking water treatment residuals as adsorbents: a review. *Minerals* 9:487. <https://doi.org/10.3390/min9080487>



- Yang Y, Tomlinson D, Kennedy S, Zhao YQ (2006a) Dewatered alum sludge: a potential adsorbent for phosphorus removal. *Water Sci Technol* 54:207–213. <https://doi.org/10.2166/wst.2006.564>
- Yang Y, Zhao YQ, Babatunde AO, Wang L, Ren YX, Han Y (2006b) Characteristics and mechanisms of phosphate adsorption on dewatered alum sludge. *Sep Purif Technol* 51:193–200. <https://doi.org/10.1016/j.seppur.2006.01.013>
- Yang Y, Zhao YQ, Kearney P (2008) Influence of ageing on the structure and phosphate adsorption capacity of dewatered alum sludge. *Chem Eng J* 145:276–284. <https://doi.org/10.1016/j.cej.2008.04.026>
- Yu-Chi L, Shang-Lien L, Jeff K, Chung-Chou T (2012) Beneficial uses of sludge from water treatment purification plants in concrete mix. *Environ Eng Sci* 29:284–289. <https://doi.org/10.1089/ees.2010.0479>
- Yusuff SM, Ong KK, Yunus WMZW, Fitrianto A, Ahmad MB (2017) Removal of methylene blue from aqueous solutions using alum sludge: sorption optimization by response surface methodology. *J Fund App Sci* 9:532–545. <https://doi.org/10.4314/jfas.v9i3s.41>
- 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 A* 45:1234–1239. <https://doi.org/10.1080/10934529.2010.493794>
- Zhao YQ, Razali M, Babatunde AO, Yang Y, Bruen M (2007) Reuse of aluminium-based water treatment sludge to immobilize a wide range of phosphorus contamination: equilibrium study with different isotherm models. *Sep Sci Technol* 42:2705–2721. <https://doi.org/10.1080/01496390701511531>
- Zhao YQ, Babatunde AO, Zhao XH, Li WC (2009) Development of alum sludge-based constructed wetland: an innovative and cost effective system for wastewater treatment. *J Environ Sci Health A* 44:827–832. <https://doi.org/10.1080/10934520902928685>
- Zhao YQ, Babatunde AO, Hu YS, Kumar JLG, Zhao XH (2011) Pilot field-scale demonstration of a novel alum sludge-based constructed wetland system for enhanced wastewater treatment. *Process Biochem* 46:278–283. <https://doi.org/10.1016/j.procbio.2010.08.023>
- 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. <https://doi.org/10.1016/j.cej.2018.07.143>
- 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* 9:612. <https://doi.org/10.3390/pr9040612>
- Zhou Y-F, Haynes RJ (2010) Water treatment sludge can be used as an adsorbent for heavy metals in wastewater streams. *Waste Manag Environ.* <https://doi.org/10.2495/WM100341>
- 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* 215:631–643. <https://doi.org/10.1007/s11270-010-0505-y>
- Zhou Y-F, Haynes RJ (2012) A comparison of water treatment sludge and red mud as adsorbents of As and Se in aqueous solution and their capacity for desorption and regeneration. *Water Air Soil Pollut* 223:5563–5573. <https://doi.org/10.1007/s11270-012-1296-0>

