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A COMPARATIVE REVIEW ON LOW-COST ADSORBENT BASED ALKALI-ACTIVATED MATERIALS BY ADSORPTION STUDY

The degradation of the condition of wastewater is becoming more and more serious due to the endless development. One of the main reasons is heavy metal contamination, which causes significant harm to the climate and humanity, such as bad health consequences, environmental degradation, and air pollution. Adsorption, which uses proven adsorbents such as activated carbon, is one of the most common methods for heavy metal removal in wastewater. However, since activated carbon is very expensive to build and repair due to complex production, most people choose another material to overcome this problem. Researchers have recently focused on finding low-cost adsorbents, which are typically industrial, agricultural and food wastes that can generate in large quantities. However, Alkali-Activated Materials (AAMs) have been recognized as a novel possible adsorbent because they are cheap, made from solid aluminosilicate and extremely alkaline activator solution, making them appropriate for usage in the civil engineering specialty. Moreover, they have become an option for various applications due to their unique geopolymer structure, which is highly mechanically, chemically and thermally stable. Hydroxyapatite (HAP) can be extremely useful in this application, as it is a promising biomaterial that has great potential for a low-cost AAMs adsorbent. The purpose of this study is to analyze the present development of a potential economic alternative adsorbent, particularly based on alkali-activated materials (known as geopolymers), for the elimination of heavy metal pollutants in wastewater using adsorption techniques.

Keywords: Adsorbent; Geopolymer; Adsorption; Hydroxyapatite; Wastewater

1. Introduction

Protecting our environment and human health is important in two ways: water quality and waste management. Rapid and continuous development around the world has led to a global increase in the accumulation of waste and the release of heavy metals into waterways [1]. This problem arises from various activities such as electroplating and chemical processing operations, manufacturing processes, household waste and service industries in various applications that are responsible for heavy metal pollution in wastewater. This phenomenon is a serious hydrological nuisance because it degrades water quality and the surrounding environment, and ultimately has a massive negative impact on human health [1,2]. Similar to waste management, if not safely disposed of, they can disturb ecological cycles, harm soils and water bodies, and produce significant air pollution such as the fog and smog that have taken place in recent years [3,4].

In view of environmental awareness and shortage of landfill space, recycling of waste/by-products as an attractive option to dispose and remove of heavy metals from wastewater becomes a priority to safeguard our ecosystem and human being.

Some of the toxic heavy metals are on the World Health Organization's list (WHO) as particularly hazardous to health, including lead (Pb), cadmium (Cd), mercury (Mg), arsenic (Ar), copper (Cu), zinc (Zn) and nickel (Ni). These heavy metals are usually found in domestic wastewater, industrial effluent, and stormwater [5,6]. These toxins penetrate the environment and produce a detrimental impact on human life and the biodiversity. Toxicity occurs because heavy metals are not biodegradable and may be carcinogenic [7-12], that may also affect the natural ecosystem. In addition, eutrophication induced by massive amounts of nutrients (phosphorus and nitrogen) in wastewater leads to a loss of dissolved oxygen, which in turn stimulates the growth of hazardous cyanobacteria [13,14]. Consequently, the existence

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of these toxins in wastewater at improper proportions might create significant health concerns for human organisms.

As response to the ecological issues presented by water contamination, many researchers had already committed themselves to the development of systems for wastewater treatment [15]. The basic purpose of wastewater treatment is to eliminate as much as possible of a polluting substance before the remaining water, called effluent, is discharged back into the environment. In past few decades, a diversity of treatment methods has been devised to eliminate harmful pollutants from wastewater. These include membranes separation, electrochemical processes, coagulation-flocculation, photocatalytic processes, advance oxidation techniques, and adsorption processes with varied degrees of efficacy to generate clean water. Among them, the adsorption process utilizing low-cost materials seems to be the most attractive as a practical and frequently used approach due to its simple design, mild operational parameters, and economic efficiency [3,16-18]. Rashid et al. [4] mentioned that adsorption is an easy, cheap, renewable and ecologically beneficial approach for wastewater treatment which is different from any other current technologies. However, efficient adsorption relies on the use of an effective adsorbent, hence further investigations are needed in this field.

Over the decades, various materials have been observed and used as adsorbents for wastewater treatment, such as activated carbon [19,20], zeolites [21], silica gel [22] and activated alumina [23,24]. Nonetheless, most of the above mentioned materials have limitations such as high cost, difficulties in regeneration, poor adsorption, and unstable at high/low pH [17,25,26]. These drawbacks have increased the demand for less expensive, yet efficient and environmentally friendly materials. In recent decades, a number of research has been conducted to investigate low-cost adsorbents for the elimination of heavy metals [27-29]. Various researchers have looked into biomass wastes, industrial byproducts, and calcium mineral wastes [30-33].

Interestingly, researchers have examined a relatively new natural substance, called as geopolimer because it has an excellent capability for the removal of contaminants through adsorption. Alkali activated materials (AAMs), sometimes referred to as geopolymers, have been widely discussed and promoted as part

of the current and future toolkit of “sustainable cement binder systems” [34,35]. AAMs have the potential to replace Ordinary Portland Cements (OPCs) due to their ability to convert waste materials with high aluminosilicate content into low CO₂ binders. In addition, AAMs have many future uses in wastewater treatment, including as adsorbents, high-pressure membranes, photocatalysts, filter media, antimicrobial media, and solidification/stabilization in residual wastewater treatment [36-38]. However, the possibility of using adsorbents based on AAMs can be a practical option for wastewater treatment compared to other applications [39]. In addition, AAM is an essential tool to enhance the economical materials because it provides a method for converting many organic and inorganic wastes into valuable materials [40-42]. AAMs can be generated in a simple and less energy-intensive procedure, commonly by purifying aluminosilicate precursors with saturated sodium hydroxide/silicate solutions at ambient temperatures, as shown in Fig 1. In addition, AAMs have an amazing physical, chemical and mechanical properties when compared to OPC, which creating this material suitable to use in various application in civil engineering, such as good mechanical properties, low shrinkage, resistance to chemical attack, excellent thermal resistance and excellent adsorption properties [34,43-46]. Therefore, it is crucial to determine the suitability of AAMs as cost-effective and environmentally friendly geopolymer adsorbents.

In this paper, the main objective is to sum up and analyze the current development of AAMs in geopolymer field as adsorbent in heavy metal removal. In addition, various types of geopolymer adsorbents, including the removal capability and factors affecting the geopolymerization process will be reviewed.

2. Low-cost adsorbent

The selection of precursor materials depends on the type of origin. Organic or inorganic materials are one of the most important criteria that can affect the properties of AAMs, also known as geopolymer. There are some studies on AAM technology that deal with the properties, possible raw materials,

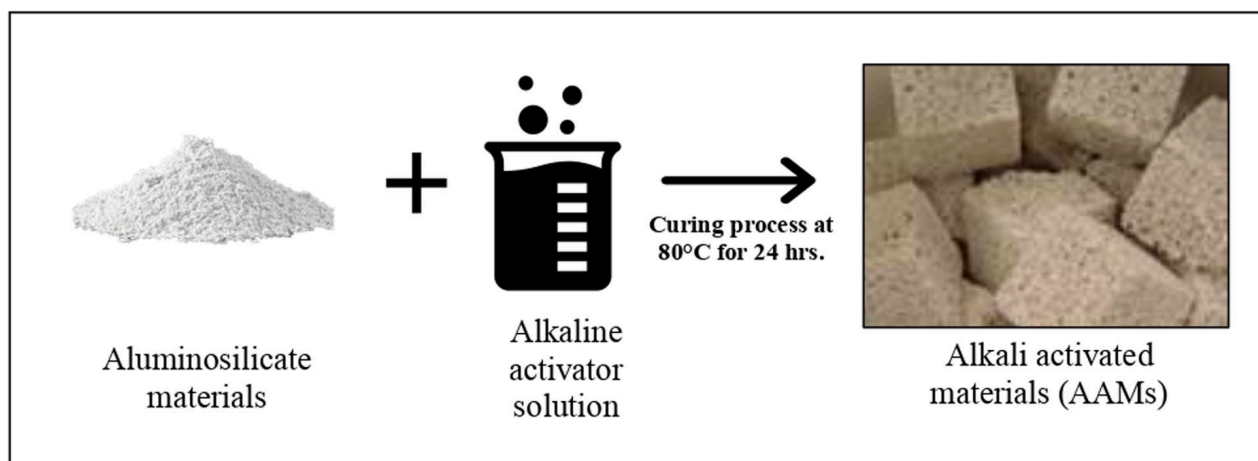


Fig. 1. Schematic diagram for production of AAMs by aluminosilicate sources created with alkali hydroxide or alkali silicate activation

chemistry and different functions of AAMs [44]. However, the purpose of this paper is to deliver a fuller overview of all recent economical AAMs applications in the field of wastewater treatment. Recently, some researchers have dedicated themselves to find AAMs adsorbents that have a larger surface area, are affordable, non-toxic, and can be easily regenerated [47,48]. To remove organic pollutants using adsorption technology, several adsorbents and their mechanisms have been explored, includes natural materials, industrial wastes, agriculture waste, bio sorbents, metal oxides, and modified geopolymer materials [49-52]. However, the most regularly used aluminosilicate raw materials as AAMs adsorbents are fly ash [53,54], metakaolin [46,55], blast furnace slag [56], and red mud [57]. Indeed, all of these raw materials may be a better option to many low-cost adsorbents, and their application will be a proven environmentally feasible and cost-effective solution [58,59].

TABLE 1 shows the current trend of aluminosilicate raw materials (low-cost adsorbent) from various studies analyzing the removal efficiency of pollutants. From TABLE 1, it can be seen that each aluminosilicate precursor has a specific ability to remove heavy metals with varying degrees of success. The study by Al Zboon et al [60] as mentioned in TABLE 1, evaluated the capability of fly ash as a geopolymer adsorbent to extract Pb (II) from aqueous solution, and the findings demonstrate that the manufactured geopolymer fly ash has greater Pb (II) removal efficiency than raw fly ash itself. It has been proven that geopolymer adsorbents outperform raw materials in terms of removal efficiency. Furthermore, by defining the optimal conditions of the parameters affecting the geopolymerization process, this study explores the adsorption properties of geopolymer and raw fly ash to remove Pb (II). Similar to a recent work by Aizat et al [61], the potential of employing dolomite in combination with fly ash as an alkaline activated material was explored while the solid/liquid ratio was controlled. The adoption of a solid/liquid (S/L) ratio of 2.5 in the mixing process resulted in promising attributes of dolomite/fly ash-based geopolymers, particularly in strength, which was 46.49 MPa, the optimal value. As a result, it was demonstrated that this composite provides an outstanding mix of

high mechanical qualities. In summary, using the proper S/L ratio results in increased strength as the workability of the alkaline activated mixer rises. As the geopolymer dosage, contact time, temperature, and initial heavy metal concentration decrease, the removal efficacy rises [46,62,63]. Furthermore, according to TABLE 1, the most common activators used in the literature for alkaline activated materials are sodium hydroxide (NaOH) and sodium silicate, as opposed to sodium metasilicate (Na_2SiO_3). Alkaline sodium-based activators are generally less expensive and have high reactivity in this context [64]. In summary, TABLE 1 shows that each type of aluminosilicate has a different effect in the composite and primarily improves the mechanical properties of the geopolymer, with various geopolymerization parameters playing a role. In recent decades, fly ash has proven to be a useful adsorbent serving as a matrix in aluminosilicate materials to remove various types of harmful heavy metals from wastewater. However, there are no study yet using hydroxyapatite and dolomite as matrix in AAMs adsorbent for removal heavy metal. Therefore, it may be beneficial to conduct the study using hydroxyapatite and dolomite as AAMs adsorbents since these materials are cheap and abundant worldwide.

3. Hydroxyapatite and dolomite as AAMs Adsorbent

The type of aluminosilicate utilized considerably influences AAMs performance [69]. Depending on whether the aluminosilicate precursor is employed alone or in combination with additional materials, AAMs are classified as single or double. The fact that most precursor materials are physically disordered, i.e. glassy (fly ash and blast furnace slag), and with thermally fractured multilayer structures complicates AAM even more (metakaolin and other calcined clays) [37,70,71]. However, some studies have discovered that adding a minor amount of calcium-containing material to the geopolymer can improve the characteristics of AAMs by up to 40% [54]. According to Canfield et al [72], increased calcium content in geopolymer adsorbent also resulted in faster set-up times, higher compressive strength,

TABLE 1

Many researchers are currently developing AAMs for heavy metal removal

Aluminosilicate precursor	Alkaline activator	Curing condition		Adsorbate	Effect in composite	Reference
		T, (°C)	T, (days)			
Fly ash	14M NaOH	105	3	Pb^{2+}	Have high mechanical strength	[60]
Metakaolin	10M NaOH & Na_2SiO_3	23	7	Pb^{2+} , Cu^{2+} Mn^{2+} , Co^{2+}	High mechanical strength and the final product contains polymeric model	[60]
		80	2			[65]
Blast furnace slag	10M NaOH	105	3	Ni^{2+} , As^{2+} , Sb^{2+}	Mortar has an excellent mechanical strength, especially when activated with $\text{Na}_2\text{SiO}_3 \cdot n\text{H}_2\text{O} + \text{NaOH}$	[66]
Fly ash/dolomite	8.3M NaOH	80	7	NR	Early strength and strength characteristics are outstanding	[67]
Coal fly ash	14M NaOH	95	3	Cd^{2+}	High mechanical properties, less energy consumption, low gas emission	[62]
Hydroxyapatite/metakaolin	8M NaOH & Na_2SiO_3	NR	NR	Pb^{2+}	Good chemical stability, ion exchange capacity, good adsorption capability, less solubility	[68]

* NR = not reported, NaOH = sodium hydroxide, Na_2SiO_3 = sodium metasilicate

and more product formation. Hydroxyapatite (HAP) is one of the materials belonging to the category of calcium phosphate compounds compatible with the general formula $\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$ that is known as major mineral component of bone and teeth. As illustrated in Fig. 2, the HAP framework is a compact assemblage of tetrahedral PO_4 groups, with each PO_4 tetrahedron shared by one column and delimiting two types of unconnected channels. Because of its extraordinary structure and intrinsic properties, this bio-inspired material has attracted the curiosity of numerous industries. According to certain studies, HAP could be a novel promising adsorbent for heavy metal removal in wastewater treatment [68]. This is due to HAP has a great thermal and chemical stability, acid-base characteristics, low solubility, high adsorption capacity, exceptional biocompatibility, environmental friendliness, low-cost material, and abundance of hydroxyl groups. As a result, HAP can be an essential constituent in geopolymer adsorbents used in wastewater treatment to extract hazardous contaminants. Furthermore, HAP can regard as an environmentally benign material in geopolymer adsorbent for a variety of reasons, including its non-toxicity and biocompatibility.

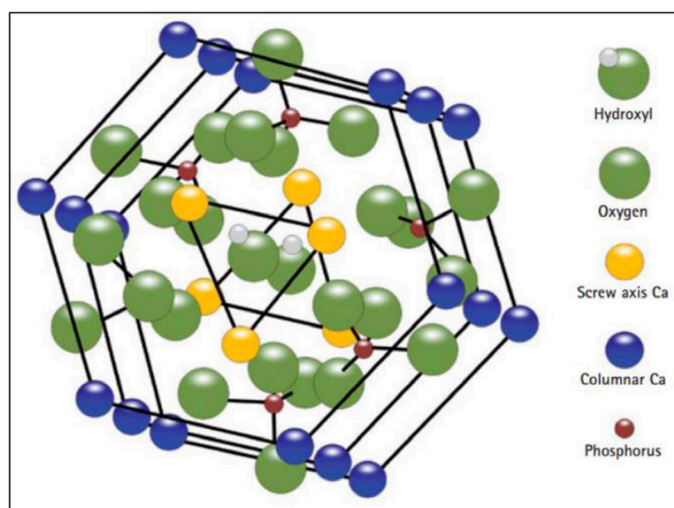


Fig. 2. The diagrammatic of the structure hydroxyapatite (HAP) which is naturally occurring form of the mineral calcium apatite consists of calcium, phosphate and oxygen [73]

Numerous studies have been conducted to investigate the use of HAP as an environmentally adsorbent due to its exceptional ability to remove various heavy metal ions, radio nuclides, organic pollutants, and dyes from wastewater [74-76]. In terms of heavy metal removal from wastewater, numerous literature studies have found that HAP have good removal capacities for various metal ions from wastewater including Pb, Zn, Cu and Cd. Gibert et al. [74] synthesized HAP via wet chemical precipitation coated with calcite particles and successfully removed Cu (q_{\max} 60.24 mg/g) and Zn (q_{\max} 34.97 mg/g) ions from water. Similarly, Choi [77] used HAP with cellulose composite in the form of a pallet to remove heavy metals. As a result, when compared to Cu, Zn, and Cd, Pb ions had the highest removal rate (up to 90% of removal). According to the findings of this study, HAP/cellulose composite is an environmentally friendly element

that solves the problem of HAP formability while maintaining superior heavy metal removal capability. From various study showed that HAP has a great potential to be an excellent AAMs adsorbent for the treatment of heavy metal ions.

Much research has been conducted to create affordable and simple removal procedures using geopolymer adsorbent materials built on carbonate minerals for the elimination of hazardous contaminants in wastewater treatment [15,78]. This has prompted several researchers to look for less expensive adsorbents, such as dolomite. This is because dolomite ($\text{CaMg}(\text{CO}_3)_2$) is a fairly cheap and plentiful material that can be obtained all over the country. According to another researchers, a recent study indicated that dolomite also functions as an effective metal sorbent. Dolomite has sparked a lot of interest because of its capability to extract specific harmful materials in a variety of applications, including heavy metals, acidic gases, de-icing salt (NaCl), and dyes, which has resulted in a slew of research studies looking into its diverse applications for treatment of wastewater.

A dolomite/HAP composite was identified in this review as a new possible precursor in the alkaline activation process to manufacture geopolymer adsorbent material. Because of its alumina, silica, and calcium content, dolomite has the potential to be used as a basic material in geopolymer as shown in TABLE 2. Furthermore, calcium compounds including dolomite and HAP are common and relatively inexpensive natural minerals that can be combined to make a low-cost adsorbent. Dolomite was employed exclusively as a filler and substitute for other raw materials in alkali-activated composites and geopolymer composites in a previous study. However, no studies have been conducted using dolomite as the primary basic material in geopolymer composites. The usage of substantial volumes of calcium carbonate of dolomite and calcium phosphate of HAP to improve the characteristics of AAMs in the geopolymer industry for wastewater treatment will be of great importance. The next section deals with the parameters affecting the geopolymerization process, which are useful for the geopolymer adsorbents to remove harmful pollutants, especially in wastewater.

TABLE 2

Dolomite's chemical composition [79]

Chemical compound	Composition of dolomite, (wt %)
CaO	80.21
Al_2O_3	1.52
SiO_2	2.50
Fe_2O_3	0.15
MgO	15.50
CuO	0.07
MnO	0.02

4. Parameters affecting geopolymerization process

Geopolymerization is an exothermic polycondensation reaction that results in the formation of an amorphous alkali activated compound known as a geopolymer from aluminosilicate

precursors and alkaline activators. The geopolymer is formed by the reaction of two components, the alkaline activator and the reactive aluminosilicate precursors. Any pozzolanic compound or silica and alumina source that dissolves readily in alkaline activator acts as a source of geopolymer precursor species and is thus suitable for geopolymerization process. The type of alkaline activator employed in the geopolymerization process is crucial. The most often employed alkaline activators were sodium hydroxide (NaOH) and sodium silicate which namely as sodium metasilicate (Na_2SiO_3) [80,81]. Furthermore, geopolymer mix designs are classified as solid/liquid ratios (S:L) and Na_2SiO_3 :NaOH ratios. Otherwise, the composition and curing procedures of a geopolymer determine its structural and mechanical properties, which include the amount of NaOH, pH, temperature, and Si/Al ratios. [82,83]. According to Panda et al [58], found that optimizing the pH, temperature, solid/liquid ratio, and molarity of NaOH resulted in the highest heavy metal removal efficiency of 98-99%. It was discovered that all of these parameters can speed up the pozzolanic reactions, which influence the final properties of the geopolymer adsorbent in terms of removing various types of hazardous heavy metals.

The solid/liquid ratio (S/L) is the aluminosilicate source/alkaline activator ratio [84,85]. The alkaline activator of the aluminosilicate source is a mixture of solid and liquid. The liquid is strongly alkaline, and the solid contains a significant amount of highly reactive silica aluminate. Alonso et al. [86] discovered that the initial solid content has a substantial effect on the rate of geopolymer production and that when the S:L ratio grew, a huge number of precipitates were detected. This is due to the high concentration of solute reactants. Because of the limited amount of alkaline activator, the geopolymer sample became less homogenous as the S:L ratio grew. Joshi and Kadu [63] used fly ash-based geopolymers to investigate the effect of S:L ratio (1.75 to 3.0) on compressive strength. According to the results of the experiments, the S:L ratio influenced the setting time and compressive strength of geopolymer in order to have good mechanical properties. For geopolymer adsorbents, the S:L ratio also plays an important role in becoming an excellent adsorbent for the removal of heavy metals in wastewater treatment. Ariffin et al. [87] studied the performance of geopolymer adsorbents based on fly ash, kaolin, and sewage sludge to determine the Cu removal from wastewater under variations of the S:L ratio. From the result, it can be concluded that different materials with different S:L ratios showed the best optimum S:L ratio with the highest Cu removal efficiency of 1.0, 0.5, and 0.8 for fly ash, kaolin, and sludge-based geopolymer adsorbents, respectively. It may be stated that the S:L ratio is vital in producing a good geopolymer adsorbent, which results in improved adsorption performance.

The concentration of NaOH (molarity) is an important factor in determining the properties of geopolymers. A previous study found that the concentration of NaOH in geopolymer composites can affect their properties and strength. Using calcined clay, Hafid et al [88] investigated the effects of NaOH concentrations ranging from 4-14M on the properties of cured geopolymer. It was discovered that the concentration of NaOH had a signifi-

cant impact on the strength of geopolymer composites. It was also reported that the flexural strength of the composites increased as the molarity increased from 4M to 11.5M and decreased as the concentration increased from 11.5M to 14M. Excess Na^+ was produced as a result of the high NaOH concentration (14M) and dissolution, which is undesirable in the polycondensation process because it weakens the structure and reduces the overall strength of the geopolymer. At the optimal NaOH concentration of 22M, Azimi et al [38] discovered that geopolymer-based dolomite/fly ash had the highest compressive strength of 46.38 MPa. In addition, the study by Hu et al. [89] investigated the effect of NaOH concentration on the adsorption of heavy metals such as Cd, Pb and Ni using a sewage sludge-based geopolymer adsorbent. Based on the experimental work, it was proved that the removal efficiency of Cd, Pb and Ni showed an increasing trend when the NaOH concentration increased from 0.25M to 7.5M. According to the IR spectral data, the adsorption effect of the bio-sorbent for heavy metals was mainly due to the complexation of N-H groups and COOH groups. Therefore, it can be concluded that NaOH concentration affects the performance of the geopolymer adsorbent by reaching the optimum concentration value, which leads to higher adsorption performance.

When considering the potential of dolomite to become AAMs adsorbent, only a few studies seem to have looked into the influence of NaOH concentration on dolomite-based geopolymers [38]. Previous research has indicated that dolomite is an excellent filler and blending material for geopolymer adsorbents [90]. However, the uses of dolomite as an important component in a geopolymer system is a new endeavor. In terms of mixing parameters and reactivity, there is also a scarcity of information on dolomite-based geopolymers [91]. As a result, it is advantageous to conduct the research to determine the optimal parameters for geopolymerization with respect to dolomite as a new precursor material.

Many researchers also investigated the ability of HAP as a geopolymer by controlling the crucial parameters of geopolymerization, including the ratio of solid to liquid, the ratio of alkaline activator, the molarity of NaOH, and the curing conditions [92,93] for mechanical performance. The research of Sukri et al. [94] revealed that the improvement of mechanical properties of HAP was achieved when the sintering temperature reached 900°C and the lower use of alkaline activator was 1.5. Furthermore, by mixing HAP with other materials to make a suitable composite material, the mechanical properties of HAP were improved. Sutthi et al. [95] pointed out that HAP together with calcined kaolin reaches maximum compressive strength (up to 37.8 MPa) when the optimum curing temperature and time are 80°C and 28 days, respectively. It was proved that the mechanical properties of HAP were improved when optimal conditions were achieved in the geopolymerization process especially when mixed with other materials. However, there is a lack of information on HAP as a geopolymer adsorbent for heavy metal removal in wastewater treatment. Only a few studies have been conducted to evaluate the capabilities of HAP as an improved adsorbent material. According to Nayak et al. [96], the features

of HAP as an adsorbent material were discussed in the context of its performance for the removal of metal ions pollutants from multi-metal ions solutions, which included Cd, Pb, Zn, Sr, and As. Thus, future research should concentrate on evaluating the effectiveness of HAP as an AAMs adsorbent for metal ion removal by controlling the parameter of geopolymerization process, as this material is non-toxic in nature, biodegradable, and widely available around the world.

5. Summary

During the last decade, there has been a significant increase in the number of studies on wastewater treatment with AAMs technology. One of the main reasons for this significance is that AAMs have many valuable properties similar to conventional materials (ceramics, synthetic zeolites, or modified polymers), such as good mechanical strength, excellent durability, porosity, ion-exchange capacity, and finally the manufacturing process is straightforward and less energy consumption. Furthermore, AAMs can be made from a variety of industrial byproducts, making them compatible with the circular economy. In addition, this review deals with the application of adsorption studies since this process promises simple operation, cost effective and universal nature among various wastewater treatment methods. Furthermore, the parameter related with geopolymerization process were discussed and concluded that solid to liquid ratio, alkaline activator ratio and concentration of NaOH are one of the most important parameters for this process. The addition of HAP during the production of geopolymer can help to increase the effectiveness of the AAMs adsorbent. However, there is a little knowledge and study related to the usage of dolomite together with HAP which should be investigated in future as these waste materials is cost effective and has a potential to be the new raw material in an alkaline activated process.

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