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Copper and Zinc Removal from Wastewater Using Alum Sludge Recovered from Water Treatment Plant

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Abstract: The study aimed to determine Aluminum sludge composition and structure for its valorisation as an alternative natural material for heavy metals removal from wastewater for further reuse as treated water in different applications. The study was conducted to investigate the introduction of Al-bearing sludge composition. The physical and chemical properties were examined using X-ray diffraction tests (XRD), scanning electron microscope tests (SEM), Fourier-transform infrared tests (FTIR), and Brunauer-Emmett-Teller tests (BET). Furthermore, the heavy metal concentrations of synthetic wastewater were measured using the spectrophotometry method. The experimental procedure is based on testing different pH limits and amounts of aluminum sludge to find the optimum conditions for copper (Cu) and zinc (Zn) removal. The results demonstrated a high removal efficiency where its value reached up to 97.4% and 96.6% for Zn and Cu, respectively, in an acidic medium (pH = 6) using a relatively high amount of sludge (1400 mg). Nevertheless, a low efficiency was obtained in the strongly acidic medium (pH = 4) and a smaller sludge amount of about 480 mg.

Keywords: aluminum sludge; heavy metals; zinc; copper; wastewater; reuse treatment

1. Introduction

Currently, the contamination of water by heavy metals is a great environmental menace that can have an impact on individuals, animals, and the environment [1]. Wastewater from various industries such as steel and mining contains high levels of heavy metals, including copper and zinc [2]. This has resulted in an accumulation of heavy metals in aquifers, which makes the precious water resources under the threat of several hazards [3]. Whereas the direct discharge of wastewaters into the sewerage systems may negatively impact the subsequent biological wastewater treatment [2]. Consequently, heavy metal pollution is one of the major environmental problems, its contamination has dangerous effects on wild and human life. Many methods have been widely used to remove heavy metal ions.

However, most of these techniques have inconveniences such as high costs, insufficient removal yield, and production of large amounts of sludge [2]. For a significant instance, laboratories are one of the sources of this type of pollution, whether within the industrial sector or scientific research activities [4]. Furthermore, the illegal methods to dispose of waste reagents with wastewater in the environment will cause further pollution [4].

Protecting the environment requires the early elimination of these metals, considering the guidelines suggested by the world health organisation (WHO) to discharge sludges into the wild [5]. Most of the heavy metal's properties (carcinogenic and teratogenic) have been considered a danger to human lives [6,7].

Among the developed viable technologies for removing heavy metals from wastewater effluents is the sorption process as a promising and highly efficient method [8].

Unfortunately, most traditional adsorption systems are broadly applied the activated carbon to remove a variety of contaminants. Despite the disadvantages related to the cost of production, separation difficulties and regeneration is still in wastewater treatment applications [9]. Standards and traditional methods of physical and chemical tests have been used to control the pollution issue, however, these tests are extremely costly. Therefore, research towards treatment processes using less costly and widely existing natural materials is promoted. Certainly, the performance and effectiveness of these techniques depend on the nature of the adsorbent, cost, abundance, and regeneration possibilities [5].

The application of Alum sludge for the removal of heavy metals could be identified as a sustainable management solution [2]. Alum sludge results from water treatment processes of drinking water treatment plants, where it is formed by adding aluminum salts to raw water to remove colloidal particles, silt and clay-size particles, color, and turbidity. Appropriately, Water Treatment Sludge (WTS) consists of particles that settle as a result of coagulation and flocculation processes [1,10,11].

Currently, it is a material deposited daily in large quantities in landfills in numerous developing countries of the world [12]. About 1–3% of the volume of raw water used in the water treatment process is produced as sludge [3,13]. Moreover, it is discharged directly downstream of the rivers or disposed of not far away from streams which in the end meet the downstream river. Both water quality and ecology are affected by such practices [14], while direct attention is necessary for the economic and respectful environmental management of alum sludge. In Algeria, Alum sludge is disposed of in sewers and landfills because of being considered a non-dangerous waste product [15].

Scientists are searching for techniques to decrease the excessive disposal of waste such as Alum sludge, which can be the cause of contamination of the receiving water bodies due to harmful chemical elements [12,16].

Alum sludge is used for the removal of heavy metals from wastewater. In fact, this method is cheaper than other methods.

Assessments have been describing and identifying several efforts to reuse aluminum sludge and future challenges in different applications. Among these applications, as specified by Babatunde, A. O. et al. [12], four categories of re-using aluminum sludge include: wastewater treatment processes, building and construction materials, structural soil, and diverse uses. While Dassanayake et al. [17] indicated four areas for reutilization of aluminum Sludge, including pollutant removal, environmental uses, agricultural sector, and industrial utilisations.

Alum sludge is used to recover aluminum [18] and is used in electrolysis with carbon-silver electrodes [18]. Ooi et al. [19] used Response Surface Methodology to obtain optimal responses in recovering aluminum, D. P. Ruziqna et al. [20] used acidification as a coagulant for tertiary wastewater treatment using Ultrafiltration [21], as an efficient sorbent for hydrogen sulfide removal H_2S in wastewater treatment plant [22]. Using Alum sludge to synthesise Zeolite LTA depending on its source [23], for phosphorus adsorption [24,25], Zeolite adsorbent from alum sludge for textile wastewater treatment [26].

Although previous studies used Alum sludge as heavy metals adsorbent, they used huge quantities and a long contact time. Where Ngatenah et al [27], showed high removal efficiency of Cu and Zn but used 2000 mg/L of Alum sludge and contact time between 75 to 180 min.

To the best of the authors' knowledge that there is no new study on heavy metals adsorption using Alum sludge as an adsorbent conducted for the large surface area from Algerian water treatment plants which affects the adsorption of heavy metals from wastewater. The paper hypothesis that re-using Alum sludge as an industrial waste would be a cost-effective method.

The main objectives of this study were to characterise Alum sludge from drinking water supply and to examine its possibility as an adsorbent of copper and zinc from synthetic wastewater. When prepared with high initial concentrations, emphasis is placed on its valorisation and reduction of waste for disposal. Most importantly, the Aluminum sulphate [$Al_2(SO_4)^3$] is the commonly coagulant used in Algerian drinking water treatment plants. As a result, this research provides an effective absorbance of Copper and Zinc removal in wastewater, to achieve an environmental and economical wastewater treatment using low-cost industrial waste.

2. Experimental

2.1. Materials and Methods

2.1.1. Sludges Sampling and Laboratory Preparation

- Sampling and conservation

The alum sludge was obtained from Bouira, a water treatment plant located in Algeria Bouira (Algiers Southeast), totaling 97 km^2. With a total population of about 108,899 inhabitants in 2017, Bouira's treatment plant has a latitude of 36°21′ North and longitude of 4°06′ East, with an altitude of 570 m, and the climate is humid there. The raw water derived to the Water Treatment Plant (WTP) from Tilesdit barrage, Figure 1 present the studied area where the sampling was collected, (WTP) uses aluminum salt as a coagulant. It has a treatment capacity of 71,208 m^3/d and 26,908,340.40 kg/year is the amount of Alum sludge generated annually which will be evacuated directly to the valley; the processes are illustrated in Figure 2.

The sludge was obtained from a settlement valve filled in bottles and conserved in a cool box at ambient temperature. At the laboratory, the sludge was dried and desiccated in the oven type Memmert model "10,003,700/387" at 105 °C for 24 h. Dried alum sludges were ground with pestle and mortar manually, sieved to 1 mm size. Finally, the sludges were conserved in room temperature to be analysed later as shown in Figure 3.

- Sludge characterisation

The Al sludge was subjected to two types of classification. First, physical and chemical sludge properties (Temperature, pH, Electrical Conductivity, total dissolved salts, and Salinity) were measured using multi-parameter (HANNA, G0068532). Also, two heavy metals Cu and Zn were analysed. The concentrations were determined using spectrophotometer (HACH lange DR6000) according to the HACH standard method.

Furthermore, the physical properties and chemical composition of the sludges were determined as follows: The Fourier transform infrared (FTIR), which is measured using IRAffinity-1 Shimadzu instrument.

FTIR scans were performed in the scanning range of 500–4000 cm^{-1}. The surface morphology and structure of the sludges were characterised using Scanning Electron Microscopy (SEM) performed on the JEOL JSM 7600F apparatus. X-ray diffraction tests (XRD) were performed on the Siemens D5000 diffractometer using CuKα1 radiation in Bragg Brentano configuration.

Surface areas were measured by employing Brunauer–Emmett–Teller (BET) on Micromeritics 3Flex analyser. The method is based on the amount of nitrogen (N2) gas adsorbed and on its surface at a specific pressure. Barrett–Joyner–Halenda (BJH) theory was applied to determine the pore characteristics (e.g., pore size, pore volume) of water sludge by referring to the N2 gas adsorption/desorption activity. For BET and BJH analyses, sludges powders were pretreated via heating (300 °C, 24 h) in a vacuum chamber to ensure the sample surface was free from water/organic vapors. The powder samples were weighed before and after the pretreatment and consequently analysed. Adsorption isotherm was recorded by measuring the total N2 gas adsorbed across a wide range of relative pressures at a constant cryogenic temperature (−196 °C). Conversely, desorption isotherm was obtained by measuring the N2 gas removal as pressure reduced.

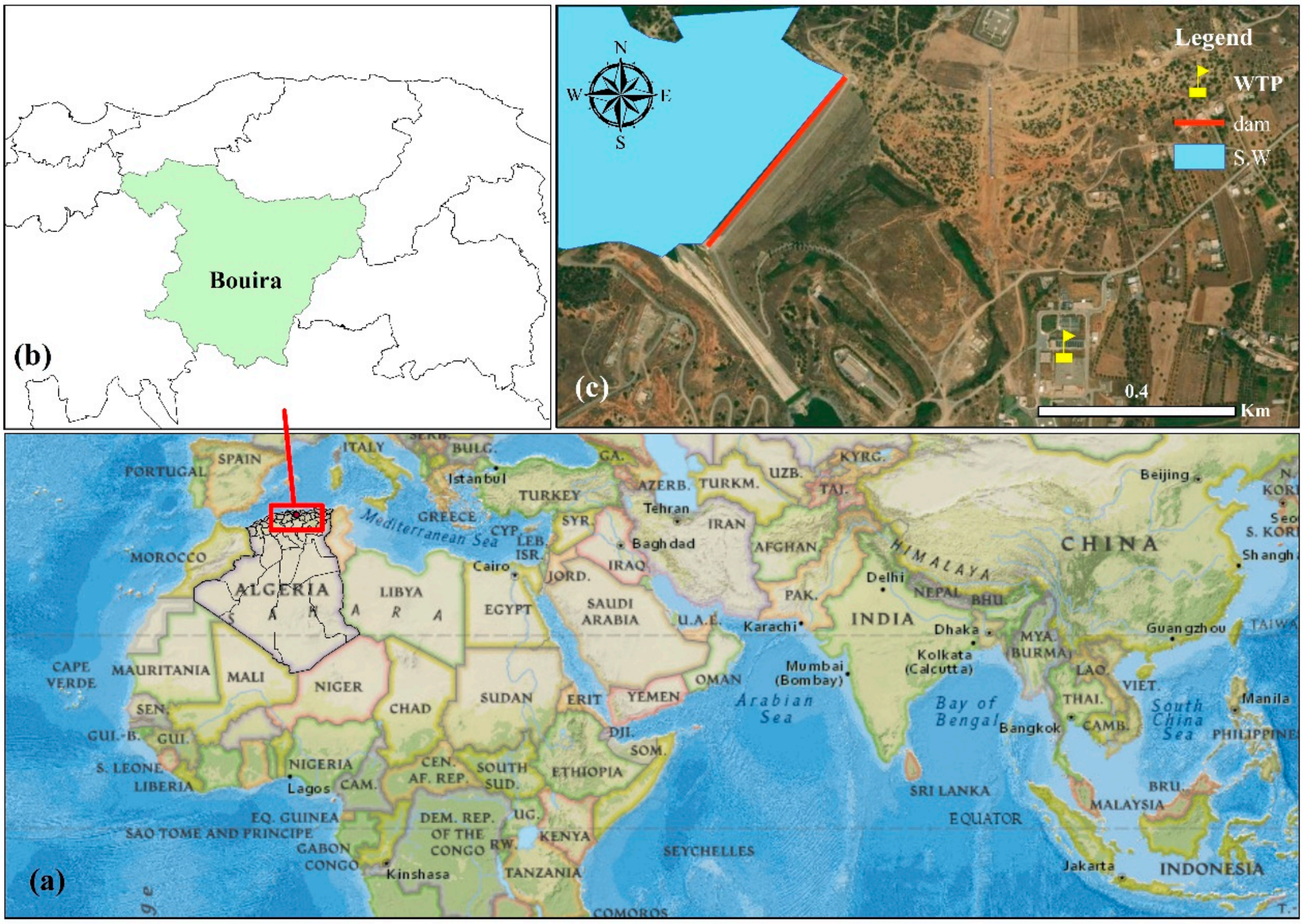

Figure 1. Map showing the sludge sampling location of the studied area. (**a**) Algeria; (**b**) Bouira city; (**c**)water treatment Plant.

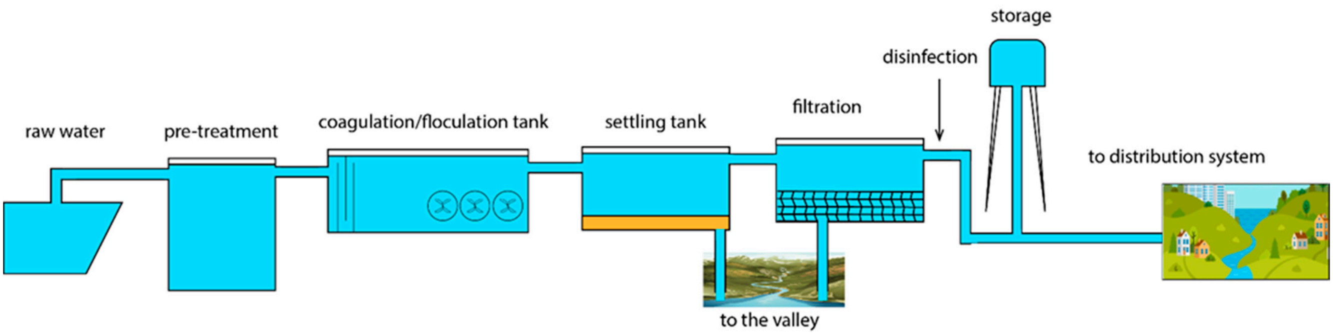

Figure 2. Drinking water treatment plant processes.

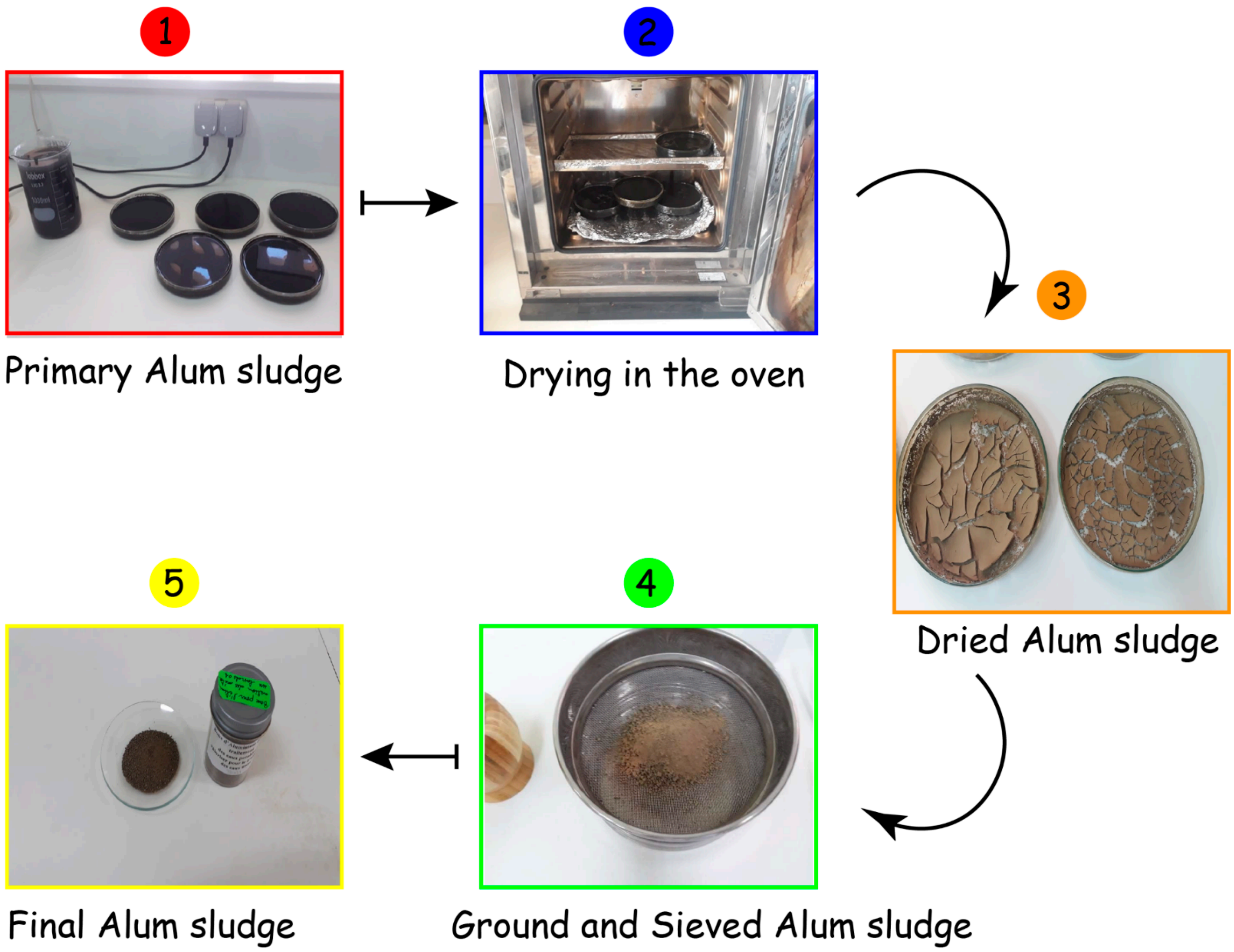

Figure 3. Schematic illustration of the preparation of Alum sludge sample.

2.1.2. Synthetic Wastewater Preparation

Synthetic wastewater solutions were prepared by dissolving copper sulfate Cu $SO_4(H_2O)_5$ and zinc sulfate Zn $SO_4(H_2O)_7$ separately in distilled water to result in known concentrations of the metal required. The anticipated concentrations of metal solutions were 5 mg/L. Different amounts of Alum sludge and the different pH ranges were made to know effectiveness of them in the removal of heavy metals. The desired pH was adjusted using HCl or NaOH, which was added to the shaker until equilibrium. All samples were shaken in a Jar test flocculator (a Stuart SW6 equipped with six rotators and six 1000 mL beakers), which was employed to simulate the adsorption process, instrument at 100 rpm for 30 min and at 200 rpm for 30 min. Then it was left for another 10 min for the sedimentation. The samples were removed and filtered using 0.45 µm pall membrane filter paper. The concentrations of filtered samples were measured using the UV-VIS-spectrophotometer (HACH Lange DR6000) according to HACH standard method. Two different procedures were conducted for each metal solution individually.

3. Results and Discussion

3.1. Physical and Chemical Characteristics of Alum Sludge

The temperature of Alum sludge was conducted at 17.20 °C, and the temperature of the area has been taken into consideration. The pH value obtained is 7.12, indicating that the sample is neutral, and this is owing to the nature and quality of the water and materials used in the treatment process.

The Electrical Conductivity range is about 0.96 dS/m, which is following the standard limits. This can be explained that the sludge is moderately mineralised for the reason of chemical products used in the coagulation process, and their concentrations. The total Dissolved Solids (TDs) is about 482 PPM under the WHO standard limits. The salinity is in the range of about 0.48 PSU. Heavy metals analyses are Copper's concentration was around 0.324 mg/L, which is lower than the standards limit of the ASCE as cited in Dassanayake et al. [17]. The main source could be an anthropogenic source as industries, copper is naturally from surface water, or the pipes' corrosion, traces of Zinc are observed in sludge 1.25 mg/L. These concentrations are under the standard limits of the ASCE, and the primary source of it is indeterminate. The analysed specific surface of the sludge is 1,767,934 m^2/g, and the specific surface area of the sludge is extremely higher. Other information regarding the sludges' pore size and pore volume the pore size and Pore volume of the sludges is also calculated. Based on these results discussed, the sludge has a high ability of adsorption. The results are summarised in Table 1.

Table 1. General characteristics and elemental composition of sludge.

Parameters	Sludge A
Temperature (°C)	17.20
pH	7.12
Electrical Conductivity (dS/m)	0.96
TDs (PPM)	482
Salinity (PSU)	0.48
Copper (mg/g)	0.000324
Zinc (mg/g)	0.00125
Specific surface (m^2/g)	176.7934
Pore size (nm)	12.1570
Pore volume (cm^3/g)	0.522754

3.2. Characterisation of Alum Sludge

3.2.1. Morphological Characterisation

The scanning electron microscope (SEM) images of sludge presented are presented in Figure 4**,** Images represent the physical morphology of the particles and the sludge structures at high and low magnifications. The alum sludge has no well-defined shapes, without a crystalline phase in the surface structure, and no uniform. Therefore, it could conclude that the Algerian Alum Sludge is amorphous. Mazaril et al. [21], Ahmad et al. [28], and Yang et al. [29] reported the same statements.

3.2.2. Structural and FTIR Characterisation

Figure 5 displays the XRD Alum sludges' patterns which are performed to discover the morphological structure and sludges' elements [30] and which detected no broad peaks over a range of spacings between (5–19°, 2θ) in sludge A. The diffractogram indicates that sludge is inadequately ordered [31]. The data in the XRD catalogue demonstrate the presence of aluminum oxide (Al_2O_3), followed by quartz (SiO_2), aluminum Sulfate ($Al_2(SO_4)_3$), Sulfur (S), and hematite (Fe_2O_3) in sludge. The results were in line with a study by Tantawy [32]. This emphasises that the Alum sludges are inadequately prepared.

Figure 6 presents the FTIR spectra of Alum sludge at a wavelength between 500–4000 cm^{-1}. A bonded O-H stretching and bending vibrations of H-O-H are observed in the spectra [28] and C-H bond, which indicates the presence of organic matter [33], stretching vibrations of C-O, and stretching bands of O-Si-O, Si-O-Si [32].

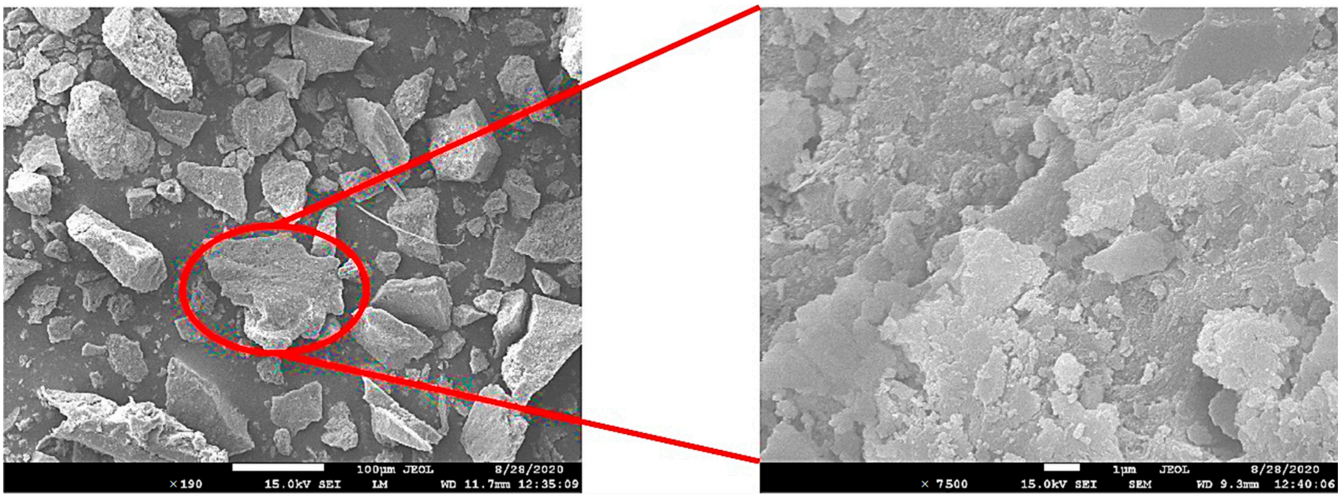

Figure 4. SEM images of dewatered aluminum Sludge.

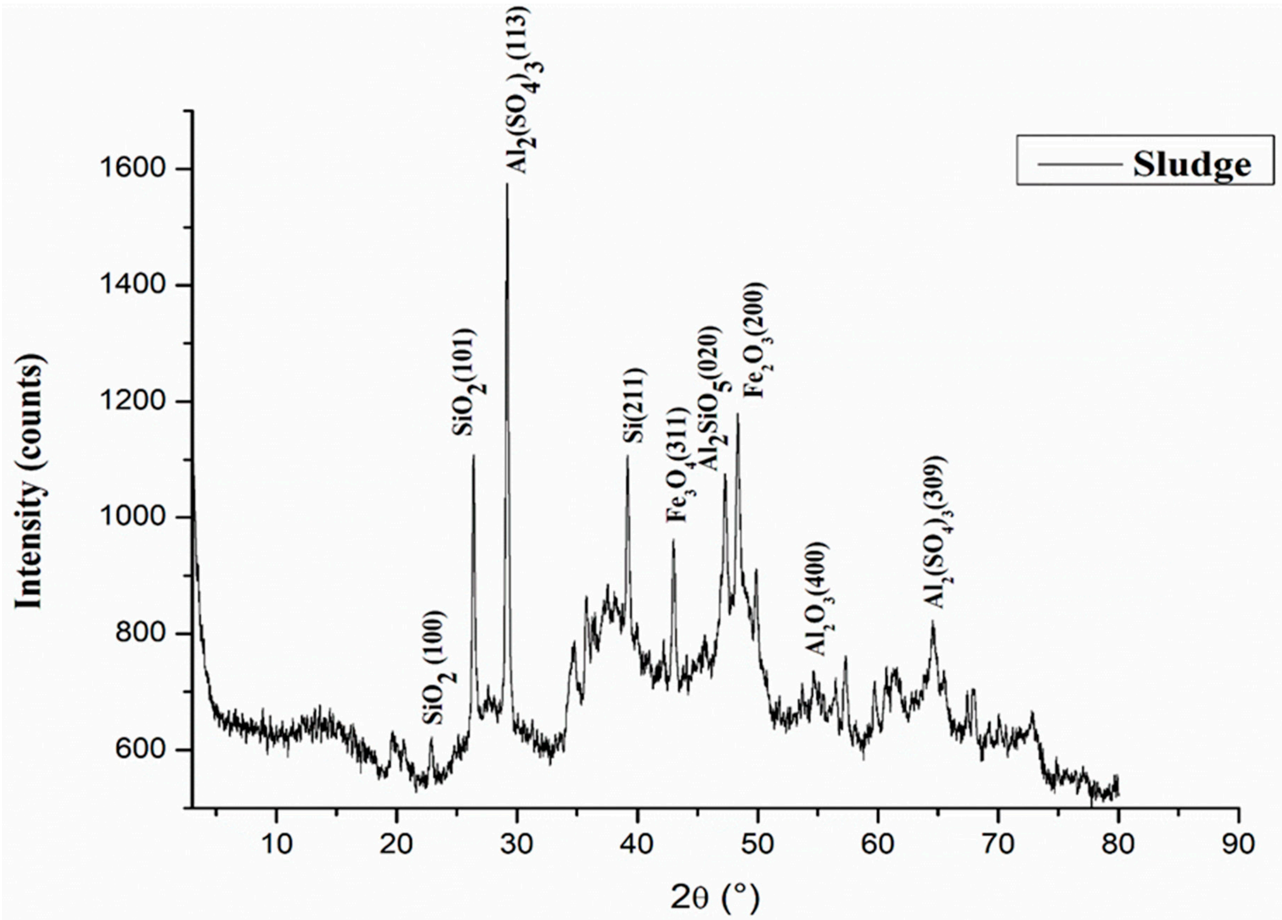

Figure 5. X-ray diffraction of Alum sludge.

Results in characterising Alum sludge re-use it in different fields and in similar areas (north of Algiers), and in comparison, it noticed that alum sludge has practically the same characterisations [21]. Mazari et al. [21] reported that Alum sludge was non-uniform and irregularly shaped for that reason it was considered amorphous, and contain a considerable amount of organic matter [21].

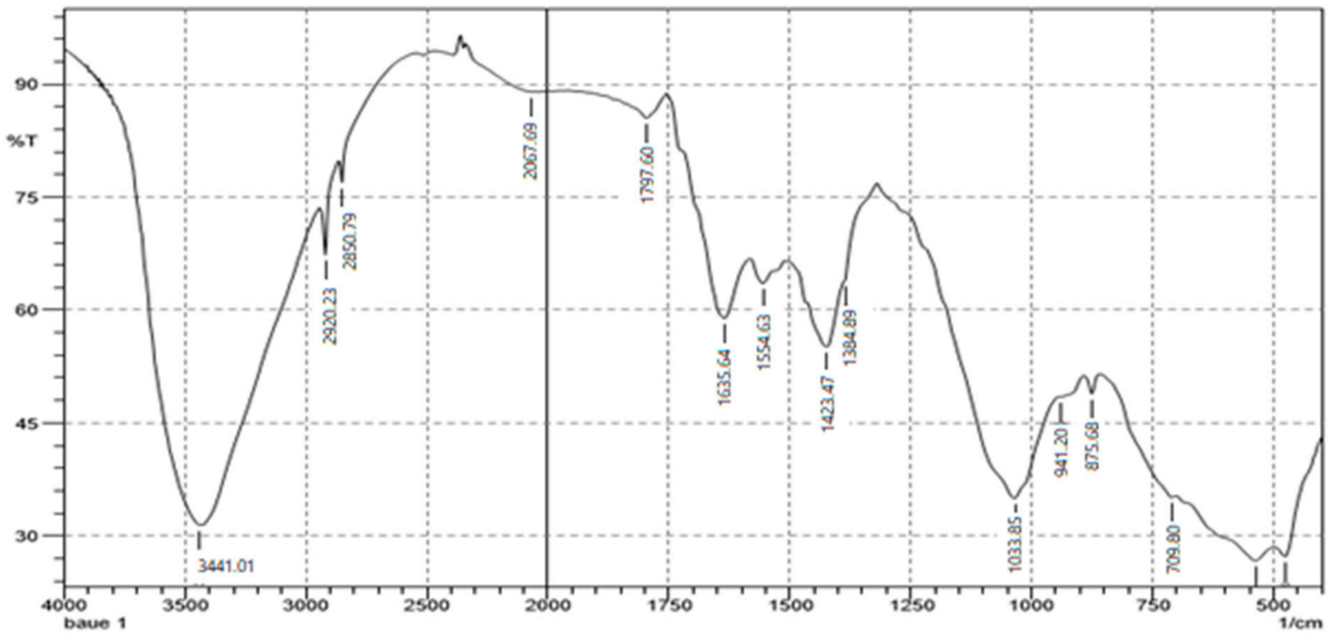

Figure 6. FTIR spectra of Alum sludge.

3.3. Efficiency of Alum Sludge on Heavy Metal Removal

(1) pH effect

The influence of pH on heavy metal removal is studied for a pH range of (4–8) as shown in Figure 7. When the amount of alum sludge is about 480 mg, the removal efficiency of Zn is increased by increasing the pH of the solution. The optimum value of pH is found to be 8, where 94.6% of Zn is removed. According to these results, Zn precipitates better at Alkaline pH where the surface of the sludge is negatively charged and thereby has a best metal removal rate. Whereas for Cu, the removal efficiency presented a different trend with higher removal of 93.6% at pH 6 and declining with an increase in pH value. Using 961 mg of alum sludge, the optimum pH was about 6 for both heavy metals with a removal of 95.8% and 95.4% for Zn and Cu, respectively. For the amount of 1400 mg, the removal efficiency revealed the same results for 961 mg.

It is known that heavy metals removal from wastewater is highly dependent on pH which has a crucial role in the adsorption process, and each heavy metal has its different optimum pH value of solubility [2].

Based on the result of Figure 6, it is noted that the pH range between (4–8), which could be effective for the removal of = Zn and Cu sludge.

(2) Sludge amount effect

To investigate the impacts of Alum sludge, different amount of alum sludge is tested for the removal of Zn and Cu from wastewater. Results are presented in Figure 8.

Figure 7 shows that the removal efficiency of Cu increases with increasing the amount of the sludge, where it reaches up to 92–96.9 and 90. This observation is probably due to the higher surface area and adsorbent sites from the amount of sludge added. According to the results, it observed that adsorption of Cu occurs at pH = 6, where the highest removal efficiency is recorded.

On the other hand, for the elimination of Zn element, Alum sludge exhibited inadequate removal in pH = 4 using 1400 mg of alum sludge, this could be attributed to overlapping and aggregating of Alum sludge sites [3], and the decrement in pH = 8 amount using 961 mg it could be due to high pH. This, results in the manifestation of a high amount of hydroxyl ions which inhibits the adsorption process of the Alum sludge [34]. A higher removal efficiency is recorded for the Zn element with about 97.4 % at pH = 6.

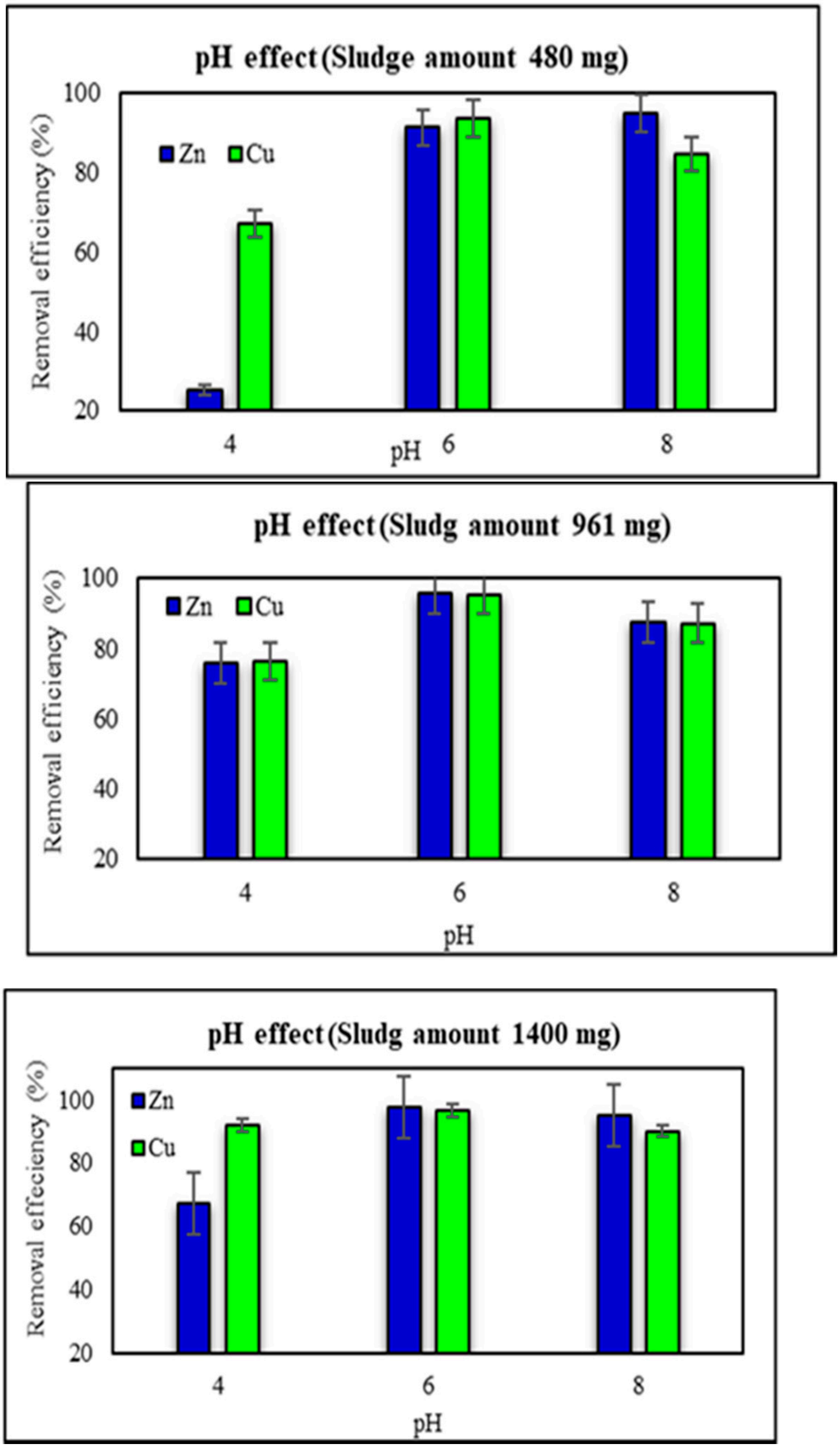

Figure 7. Effect of pH in Heavy metals removal using different sludge amounts.

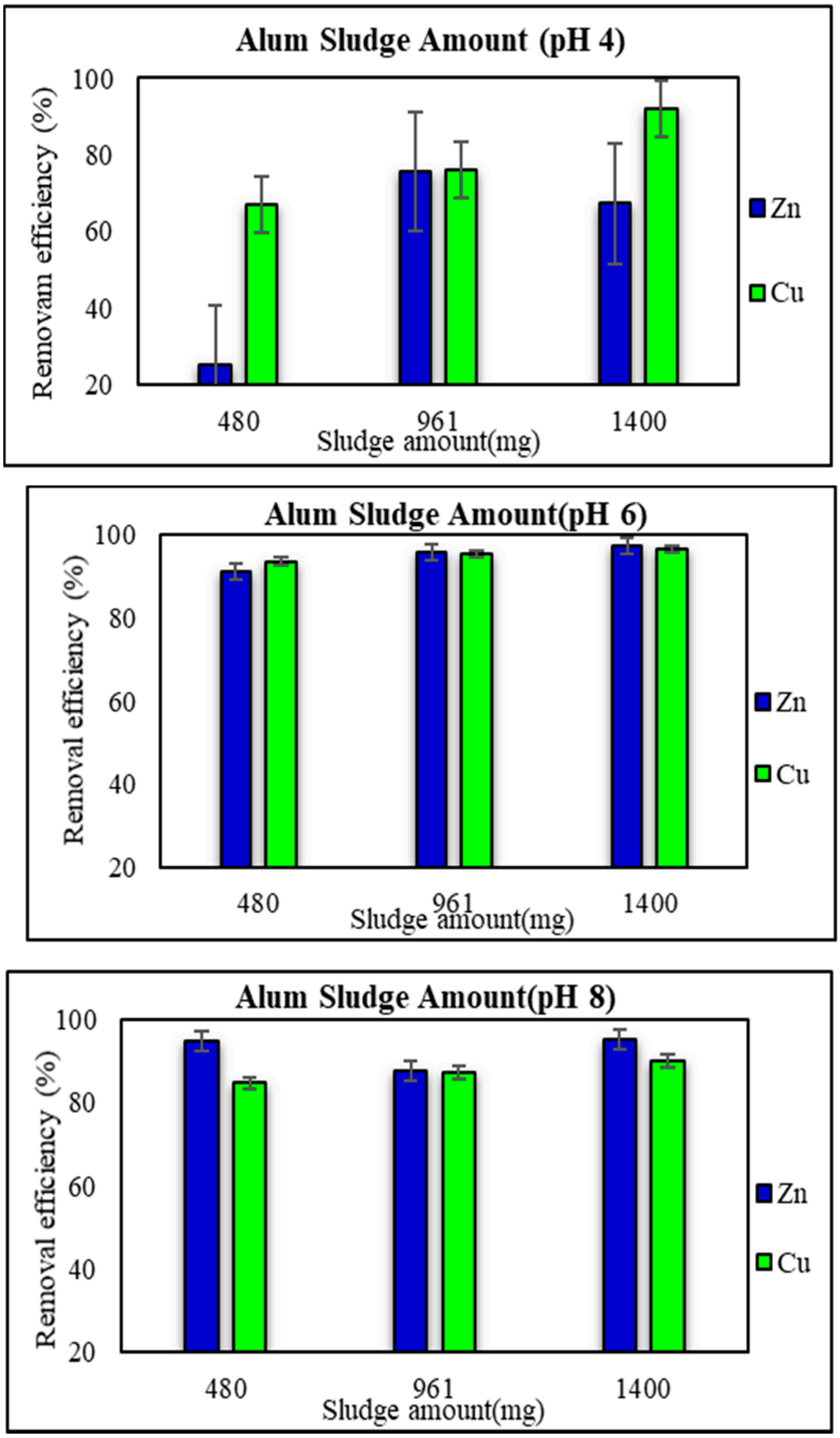

Figure 8. Effect of Sludge amount in Heavy metals removal in deferent pH mediums.

4. Conclusions

The characterization of the alum sludge demonstrates that the results of Bouira's water treatment plant and the concentration levels of heavy metals are within admissible limits. XRD results concluded the existence of major mineral kaolinite, and the sludge is inadequately ordered. The SEM results showed that the sludge is amorphous in nature. The functional groups and types of bonds present in the sludge were examined using the FTIR. Results showed that the sludge contained a considerable amount of organic matter. The sludge is highly porous in nature due to its greatest specific surface, and due to all these characteristics and properties, it could be beneficially re-used as a cost-effective adsorbent, which reduces the concentrations of Cu and Zn in synthetic wastewater.

The amount of Alum sludge and the pH are the factors that affect the removal of Zn and Cu in wastewater. Based on the results, high removal in different pH points and other Alum sludge amount have been obtained and suggested three conditions:

- A small amount of Alum sludge with pH = 4 is destined for industrial wastewater.
- An average amount of Alum sludge with pH = 6 is destined for drinking water.
- A high amount of Alum sludge with pH = 8 is destined for wastewater.

Finally, the alum sludge from Algerian water treatment plant showed to be considered an excellent suitable sorbent and industrial Algerian waste for re-use and for disposal, where the removal efficiency reached up to 97.4% and 96.6% for Zn and Cu, respectively.

Re-using Alum sludge in wastewater treatment could be encouraged for its availability in huge quantities and low cost, as such this leads authorities to test it commercially.

There are high environmental risks of heavy metals such as Copper and Zinc in water; in Algeria, municipal wastewater is evacuated directly into the environment. In future research, the factor impact of time should be taken into consideration in order to study the possibility of improving the removal efficiency.

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