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Compressive Strength of Geopolymeric Cubes Produced from Solid Wastes of Alum Industry and Drinking Water Treatment Plants

Mohammed Abdelmawla[1]*, Ahmed Abdelaal[1], Mokhtar S. Beheary[1], Nabil A.Abdullah[2], Taha M. A. Razek[3]

[1]*Environmental Sciences Dep., Faculty of Science, Port Said University, Port Said, Egypt.*

[2]*R&D Director Aluminum Sulfate Co., Egypt.*

[3]*Institute of Environmental Studies and Researches, Ain Shams University, Cairo, Egypt.*

IN this work, geopolymer is produced from two by-products waste as an alternative environmentally green construction and building materials without using Ordinary Portland Cement (OPC). Water Treatment Sludge (WTS) from Marg Drinking Water Treatment Plants in Cairo and De-Aluminated Kaolin (DAK) from Egyptian Company for Aluminum Sulfate were used in this study. Sodium hydroxide (NaOH) solution was used as an alkaline activator. The effect of the various influential factors on compressive strengths of WTS/DAK geopolymer was investigated. These factors are mixing ingredient (WTS/DAK) ratios and amounts of NaOH of different normality. The mineralogical and chemical compositions of the WTS/DAK wastes were obtained using X-ray Diffraction (XRD) and X-ray Fluorescence (XRF) analyses. Results show that the optimum ingredients providing maximum strength are the Na_2O/SiO_2 ratio of 0.56 and WTS/DAK ratio of 30:70. The development of compressive strengths over curing time of WTS/DAK geopolymer samples at optimum ingredients (30:70 WTS/DAK, 60 g NaOH 10 N and 70 oC for 72 hours). The compressive strength of WTS/DAK geopolymer gives 17 MPa after 7 days and increased to maximum strength of 22 MPa at 28 days. The compressive strengths obtained comply with the Egyptian Industrial Standards. Moreover, the WTS that traditionally disposed into landfills or drainage canals can be used sustainably in developing cement-free geopolymers with economical and environmental significance.

Keywords: Geopolymer, Water treatment sludge, dealuminated Kaolin, Compressive strength, Aluminum Sulfate, Environment.

Introduction

The concept of waste management produced from various human activities has become one of the main avenues disciplines of environmental science, especially its economic benefits. This has led to the development and improvement of waste management technologies and investment of the waste as a resource rather than disposal. Hence, the approach of waste management to profitability purpose becomes urgent [1].

Geopolymer is emerged as possible technological solution for the effective management of solid industrial wastes, as it achieves to turn a numerous of them into added value products [2, 3, 4, 5], and stabilize and/or immobilize hazardous and toxic materials [6, 7, 8, 9, 10, 11]. Geopolymers are further more touted for their high performance (high strength and durability), low CO_2 emission and low energy consumption. Silica-rich materials such as clay minerals especially meta-kaolinite are used as a precursor to react with the liquid alkaline activator [12].

According to zero-waste concept, many studies investigated recently the use of water treatment sludge as building materials [13]. Previously this sludge has been mixed with sand and cement to manufacture a cement-sludge bearing unit.

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*Corresponding author e-mail: mohamed.abdelmawla.eg@gmail.com

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However, the use of conventional portland cement will result in large emissions of carbon dioxide, dust, and heat [14]. Alkali activated alumino-silicate cement, known as 'geopolymer', has become increasingly interested in recent years as an environment friendly alternative to ordinary portland cement (OPC) [15, 16].

Production of drinking water is one of the vital industries related to human life [17]. Drinking water treatment depends on natural resources, which determine the appropriate technology such as desalination of sea water, clarify of freshwater or extraction of groundwater [18]. Clarifier system employed in water treatment plants results in the muddy sludge settled to the bottom of the treatment tank. The liquid sludge is subsequently drained to sludge lagoons or drainages for disposal. The increasing demand of treated water produced has resulted in increasing quantities of sludge by-products generated annually [19].

Recent studies investigated the use of water treatment sludge to replace cement in the production of paving tiles [20], clay bricks [21], ceramic bricks [22] concrete by mixing WTS with rice husk ash produced from agricultural waste [23] and composite cements by substituting granulated slag with fired drinking water sludge [24].

The Egyptian Holding Company for Water and Wastewater, along with 23 subsidiary companies, which established in 2004 produces huge quantities of drinking water up to 22 million m^3/day [25]. This is accompanied by liquid aluminated sludge production which is estimated at 10% of this quantity and is increasing according to the needs of the population. Hence, the urgent need to find a sustainable reuse option for growing stocks of sludge with limited beneficial usage previously disposed of in waterways or destined into landfills or drainages.

In Egypt, Handling of muddy water or sludge from WTP is done by reuse, recycling or disposal ways. WTP such as Roadelfarag reuses the rejected muddy water from the purification process, by adding to fresh water to reduce the alum dose since it has ratios of the remaining alum. Other WTP such as Sheikzaied drains the muddy water into the lagoon to evaporate the water content and recycling the dry sludge. Marg WTP uses a Belt-press unit to remove any water content and thickening then it is a good resource in multi-uses [26]. However, most of WTP is disposing of the sludge from WTP, as is, into the drainages and waterways directly. It is causing the problem with the Egyptian Environmental Affairs Agency (EEAA) and Ministry of water resources and irrigation (MWRI) [27, 28].

Aluminum sulfate is the most common coagulant used in the Water Treatment Plants which is produced in the Egyptian Aluminum Sulfate Company from kaolinite rich-in alumina which then leached by concentrated sulfuric acid to produce aluminum sulfate [29]. Previous studies investigated the possibility of using a precursor from De-aluminated meta-kaolin and WTS activated with NaOH to produce geopolymers [30, 14]. The strength requirement by Egyptian Industrial Standards is 2.5 MPa for non-bearing and 20.0 MPa for bearing building units (ASTM, 2017) [31].

The objectives of this study are investigating the strength characteristics of WTS/DAK geopolymer to ascertain its performance as a construction units using liquid alkaline activation. The influential factors include different ratios of mixing ingredients were highlighted and discussed. The structural composition of WTS/DAK geopolymer was illustrated using X-ray Diffraction (XRD) analysis to understand the role of influential factors controlling the strength development. The top outcome of this paper is enabling sludge, traditionally destined into landfills or water ways, to be used in a sustainable manner as a geopolymer masonry unit with a significant value in term of engineering, economic and environmental perspectives. The environmental aspects of the WTS/DAK geopolymer will not be discussed in this paper.

Materials and Methods

Sample preparation

WTS and DAK samples were grinded to be a fine powder and followed by grain size analysis (Fig. 1). The WTS/DAK geopolymer sample was prepared by combining two-sludge and adding NaOH as a solution alkaline activator. The WTS/DAK ratio was fixed at 50:50 as weight and different amount of NaOH concentration were 2, 4, 6, 8, 10 N respectively. Then WTS/DAK was mixed for 5 minutes in a mixer to ensure homogeneity of the mixture. The mixer was stopped and the mixture was activated by adding NaOH and mixed for additional 5 minutes. Final mixture was then molded into cubes with dimensions of 5 cm^3, whereby the compression was done using a mortar Handle. The sampled cubes were dismantled and then heated at 50, 60, 70, 80 and 90°C for time durations of 24, 48 and 72 hours respectively. After heating, the samples were subsequently cured at room temperature until lapse of different curing times as planned.

Materials and Sieve analysis

WTS samples were collected from Marg Water Treatment Plant located in Marg village in Cairo which is affiliated to the Drinking Water Company in Cairo, Egypt. The sludge consists of a ratio of alumina from the coagulation process in the Water Treatment Plant. DAK samples were obtained from the Egyptian Aluminum Sulfate Company. Sieve Analysis was carried out to indicate the grain size distribution of the studied WTS and DAK samples as shown in Fig. 1.

Mineral and chemical compositions of WTS and DAK

The mineral composition of WTS and DAK was obtained from XRD analysis (Fig. 2) using PANalytical X'Pert PRO X-Ray Diffraction equipment.

WTS and DAK samples were analyzed for the chemical composition using Philips X-ray fluorescence (XRF) spectrometer Model PW/2404 and given in Table 1. Both XRD and XRF analyses were carried out in the central laboratories of the Egyptian Mineral Resources Authority (EMRA) at Cairo, Egypt. Loss on ignition (LOI), which is a measure of percentage of organic content in the sample, was determined by oven drying 2g of material at 105°C to constant mass before calcining at 800°C for 2 hours, cooling and re-weighing. The loss in weight is figured as a percentage of the original sample [32].

Compressive Strength of WTS/DAK geopolymer cubes

Compressive Strength test is the most important test for assuring the engineering quality of a building material [22]. Compressive strengths of WTS/DAK geopolymer cubes were measured after 7, 28, 56 and 84 days of curing according to ASTM D 1633 Method [31]. The compressive test was carried out using Non-Automatic Compression Range 200 KN-Hoek Cell Machine in the Material Test Laboratory, National Research Centre at Cairo, Egypt.

Results and Discussion

Sieve Analysis was carried out to indicate the grain size distribution of the studied WTS and DAK samples as shown in Fig. 1. It is obvious from the grain size distribution that 53.73% of WTS and 87.87% DAK samples are finer than 120 µm (Fig.1).

DAK is composed mainly of amorphous phase with some peaks of crystalline phases of Quartz, Sillimanite (aluminosilicate) and Anatase (titanium dioxide) (Fig. 2A). While WTS composed of amorphous phase with some peaks of crystalline phases of Quartz, Albite and Montmorillonite (Fig. 2 B).

The ratio of silica and alumina influences the properties of the geopolymers [33]. The SiO_2/Al_2O_3 ratio was calculated from Table 1 considering that silica of WTS occurs in crystal phaseand will be subtracted in final calculations. The ratio of WTS/DAK 50:50 was achieved when SiO_2/Al_2O_3 ratio was approximately (5:1) complying with that reported by [34]. Different amounts (10, 20, 30 ..., and 100 g as a solution) of NaOH 10 N were added to 50:50 WTS/DAK sample then mixeda nd molded in cubes with volume of 5 cm^3. The compressive strength test was performed 28 day after the sample cubes cured in the room temperature. The appropriate amount of NaOH 10 N is determined in relation to the compressive strengths of WTS/DAK geopolymer cubes as shown in Fig. 3.

Sodium hydroxide solution (NaOH) dissolves silicate and aluminate from DAK and WTS which in return enhances the geopolymerization reaction. The smaller amount (e.g. 10 g) of NaOH was not enough to dissolve silicate and alumina, however the larger (e.g. 100 g) of NaOH caused micro-cracks on the particles due to strong base concentration and be fragile [35]. at 80, 90 and 100 g NaOH per WTS/DAK mixtures, the samples failed to achieve any compressive strength test due to the excess of NaOH which reacted with CO_2 in the atmospheric media and the samples became very fragile. Furthermore, samples at 10 to 60 g NaOH achieved the compressive strength test with increasing amounts of NaOH 10 N and achieved the highest strength at 60 g mixed with 50:50 WTS/DAK (Fig. 3).

The effect of normality of NaOH on the compressive strength development in the WTS/DAK geopolymer is illustrated in Fig. 4, which shows the relationship between 28-day compressive strengths against the normality of NaOH as activator for the WTS/DAK ratios of 90:10, 70:30, 50:50, 30:70 and 10:90 respectively.

TABLE 1. Chemical compositions of WTS and DAK (% by weight of dry sample).

Chemical composition	WTS(%)	DAK(%)
SiO_2	46.74	82.83
Al_2O_3	20.41	5.99
Fe_2O_3	4.75	0.5
TiO_2	0.98	3.4
MnO	0.08	0.01
MgO	0.47	0.09
CaO	2.13	0.15
Na_2O	0.10	0.03
K_2O	0.40	0.05
P_2O_5	0.12	0.01
Cl	0.15	0.06
SO_3	0.53	0.85
LOI (Loss on Ignition)	22.81	5.84
Total	99.67	99.81

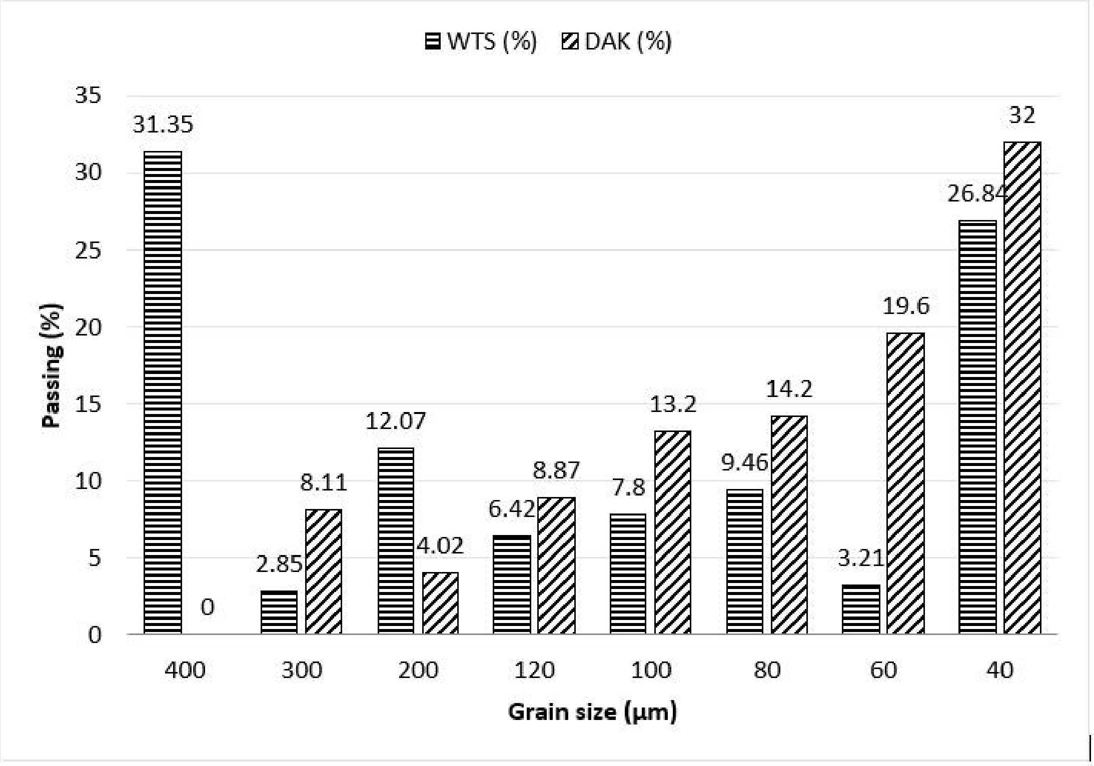

Fig. 1. Grain size distribution of WTS and DAK (by weight % of dry sample).

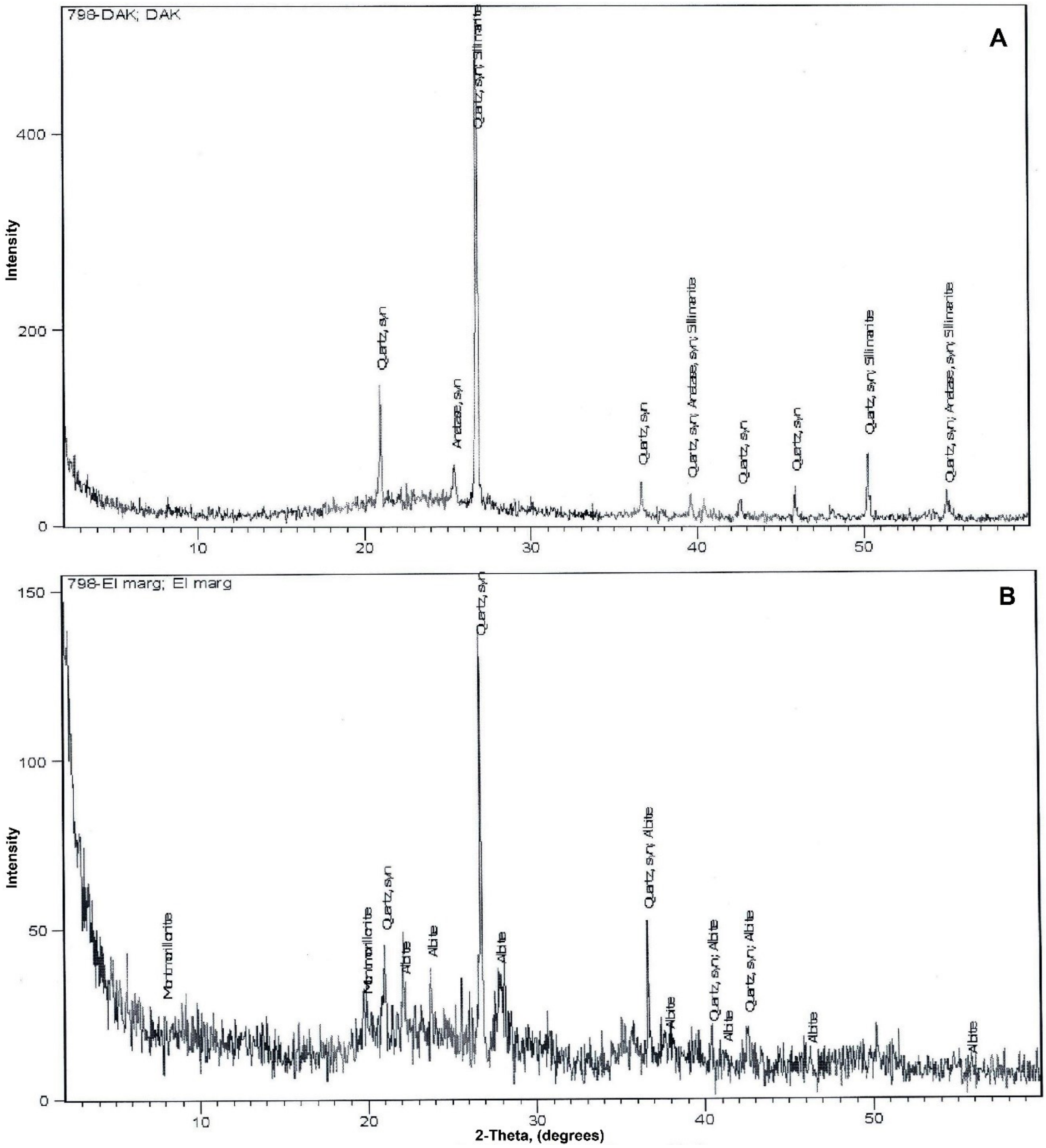

Fig. 2. XRD patterns of DAK (A) and WTS (B).

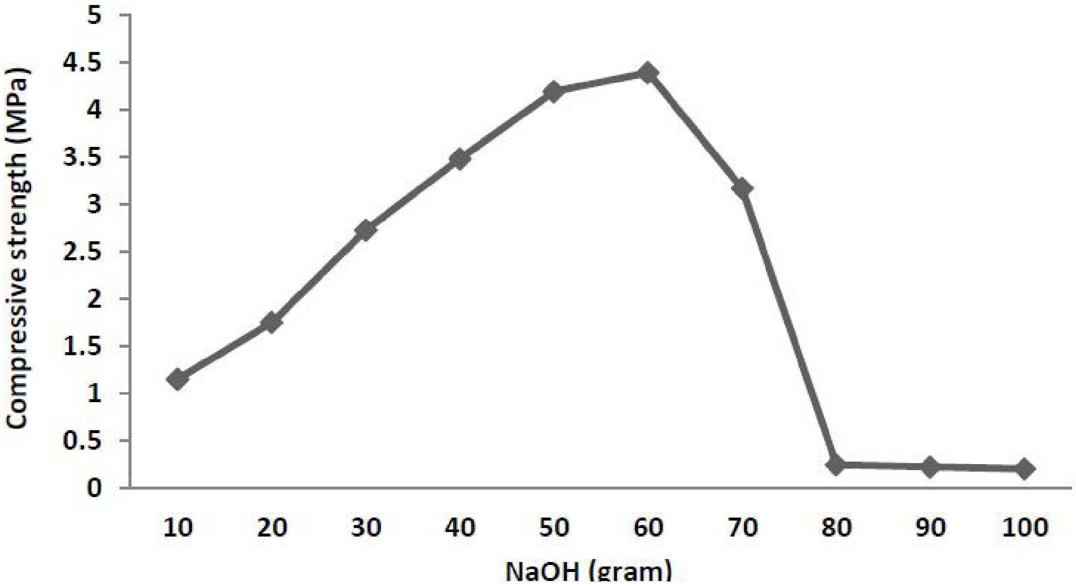

Fig. 3. Distribution of different amounts (gram) of NaOH 10 N in relation to the compressive strengths (MPa) of WTS/DAK geopolymer cubes cured in room temperature.

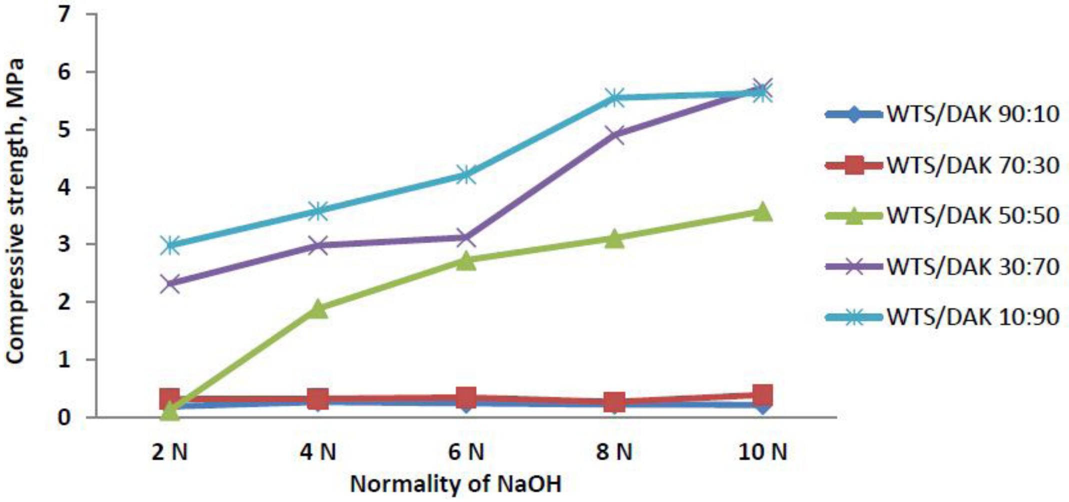

Fig. 4. Compressive strengths of geopolymer as a function of WTS/DAK mass ratios and different normality of NaOH.

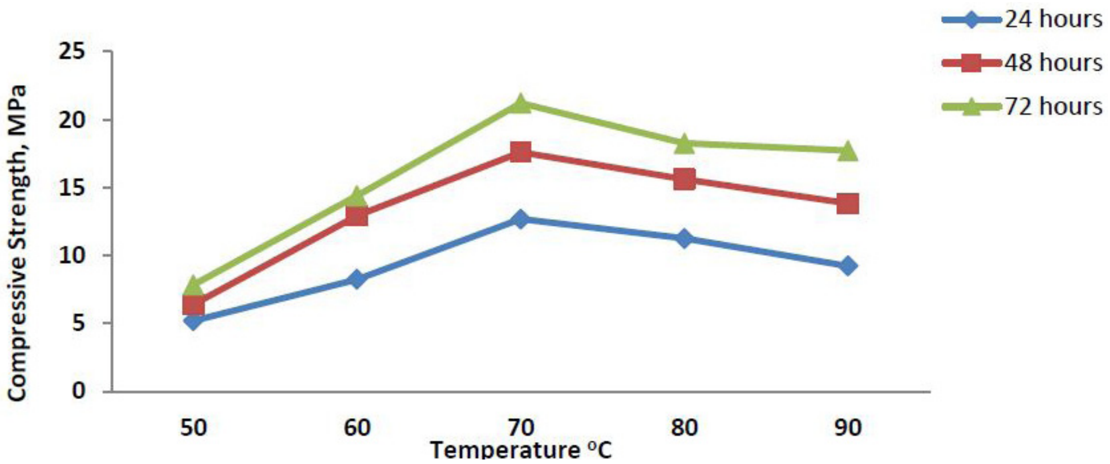

Fig. 5. 28-day compressive strengths of WTS/DAK geopolymers related to different heat temperatures.

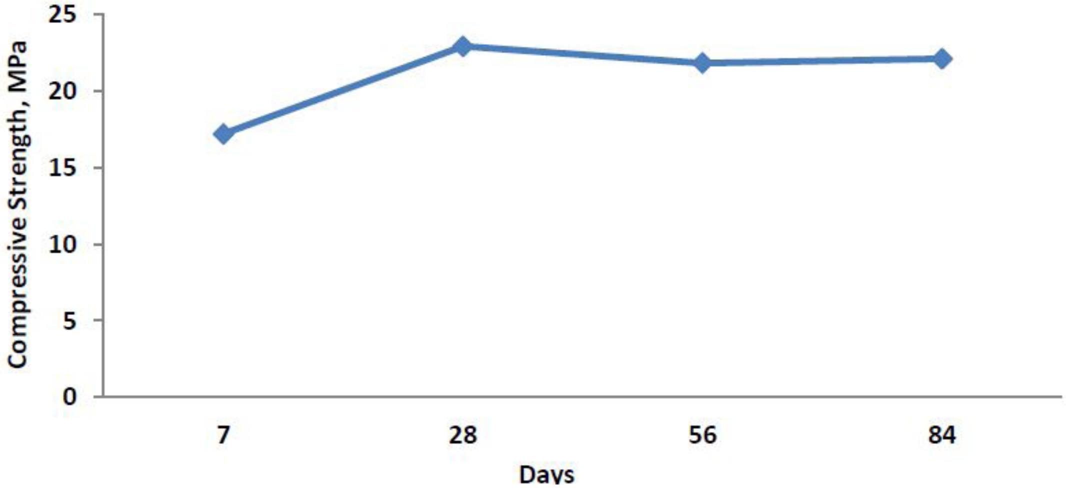

Fig. 6. Development of compressive strengths of geopolymer over curing time (days).

The samples were dried in room temperature. The 10 N of NaOH gives the maximum compressive strength for all WTS/DAK ratios [36]. Moreover, any excess of WTS showed weak strength of the geopolymer. This could be referred to the SiO_2 content in WTS which obtained from the soil in crystalline state [10], and retards the geopolymerization reaction. The test result showed that the maximum compressive strengths of WTS/DAK geopolymers are obtained at 30:70 ratio using 60 g of NaOH 10N (Fig. 4). SiO_2 and Al_2O_3 In the formation of geopolymer must be in an amorphous state.

The relationship between 28-day compressive strengths and heating duration of WTS/DAK geopolymer samples (24, 48 and 72 hours) for various heat temperatures (50, 60, 70, 80 and 90^{o}C) is shown in Fig. 5. The compressive strengths of WTS/DAK geopolymer samples increase as the heating duration increases until threshold heat duration of 72 h as curing time. The 28-day strengths of samples cured at higher temperatures are significantly higher than those cured at room temperature until 70^{o}C, indicating that the heating of WTS/DAK geopolymer samples stimulates the geopolymerization reaction. However, at a temperature greater than 80°C, some minor cracks may appear and reduce the compressive strength of other samples as a result of water loss (Fig. 5).

From Figs. 4 and 5, the optimal conditions for the WTS/DAK Geopolymer formation according to the previous parameters are 30:70 WTS/DAK using 60g of NaOH per 100 g of Mixture as an activator at 70 ° C for 72 hours [35].

The development of compressive strengths over curing time of WTS/DAK geopolymer samples at optimum ingredients (30:70 WTS/DAK, 60 g NaOH 10 N and 70^{o}C for 72 hours) is given in Fig. 6. The compressive strength of WTS/DAK geopolymer gives 17 MPa after 7 days. While the compressive strength increases until 28 days of curing and gives maximum strength of 22 MPa, which exceeds the strength requirement for masonry bearing unit (Fig. 6). Moreover, the final (long-term) compressive strength of WTS/DAK geopolymer cured at room temperature is lower than that cured at higher temperatures. The heat energy is not only accelerates the geopolymerization reaction at an early state (as noted by high early strength) but also improves the final (long-term) compressive strength.

Conclusions

1. To conclude, two solid wastes (sludge) from Drinking Water Treatment Plants (WTS) and De-aluminated kaolin (DAK) yielded from the Aluminum Sulfate Industry, were used successfully in producing geopolymer which can be used as green alternative for building materials without using Portland cement.

2. WTS that traditionally destined to landfill or into drainage canal can be used in a sustainable manner as alternative aggregates to develop geopolymer masonry units.

3. The most stable compressive strength of the WTS/DAK geopolymer developed in this study at optimum ingredient and heated at 70^{o}C is about 20 MPa with 72 hours as heat duration.

4. Furthermore, the compressive strengths of the developed WTS/DAK geopolymer comply with the Egyptian Industrial Standards.

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