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Environmental safe disposal of cement kiln dust for the production of geopolymers

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Abstract

A large amount of industrial byproducts are released from heavy industries such as power generation industry, steel industry, cement industry, etc. These wastes or byproducts like fly ash, CKD, bottom ash, blast furnace slag, methakaoline, etc. pose various difficulties in getting rid of them. May the dynamic solution is to use these byproducts for some other beneficial application.Clearly, cement industry has been found to be an energy-intensive industry that acts as a major source of greenhouse gas emissions especiallycarbon dioxide, so, with the continuous growth of construction activities, cement industry will last for a long time due to the need for high infrastructure.Recycling of industry wastes will enhance their economical and environmental values. Production of geopolymers from the wastes of cement industry (CKD) defiantly save the raw materials, and reduce the CO_2 emissions. In this work, local raw materials are used such as blast furnace slag, silica fume, CKD, meta-kaolineand alkaline activated solution of potassium hydroxide and potassium silicate has a molar ratio of K_2O:SiO_2 (0.7). The products obtained were tested for unconfined compressive strength setting time and workability, results show that it possible to produce good quality geopolymers (useful product)using industrial by-products as precursors.

Key words: Geopolymers, Industrial byproducts, CKD, and Phases Composition.

1. Introduction

Reducing global warming requires nearly zero long-term, man-made emissions.This means that, while some human works emit huge greenhouse gases to the atmosphere, others remove the same amount, leading to a net-zero balance. Limiting global warming to 2◦C requires to reach net-zero emissions by 2100 [1-2]. Consequently, coal-fired power plants beside a variety of industries, such as cement, steelmaking, and petrochemical industries, emit very large amounts of carbon dioxide that contribute to climate change through global warming of the earth. The global CO2 emission from cement industry was 2.30±0.20 gt CO_2 in 2020, which is around 8.6% of the global CO2emissions [1-3]. While the global demand for cement is expected to increase by 12% to 23% in 2100. International Energy Agency (IEA) predicts that direct CO_2 emissions from cement production will roughly increase 4.1% in the same period [4-6]. Many opportunities exist for CO_2 emission in the cement industry, with three preferred mitigationmeasures have been adapted over the last years e.g: a)_energy efficiency improvement, b)_fuel switching by use waste as alternative fuel, and c)_blended cements by reduction of clinker/cement ratio using industrial byproducts and/or sustainable raw materials[7]. The chemical composition of the collected cement kiln dust from the kiln inlet point known as the bypass opening of the vortex varies with different production line process. The concentration of lime in fresh dust ranges between 42% to 60% with an average of 51.6%. [8-10]. The maximumamount of chloride and sulfate was about 8.0% and 11.5% respectively. CKD consists of ultra-fine particles that are

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collected as mentioned above and during the production of cement clinker. The concentration of lime, sulfate, and alkali in CKD is primarily influenced by the particle size of the materials collected at the kiln inlet [11]. Coarse/heavy particles of CKD have a high content of lime while fine/light particles have a higher concentration of sulfates and alkalis with a lower lime content [12-13].

To reduce the dust emissions from cement industry chimneys, CO_2 emitted throughcalcination can be captured and utilized either by mineral carbonation of industrial solid waste or geopolymerization process using alkaline activated solution. The carbonated materials maythen be used as fillers for blended cement production [14-15] or can replace OPC in some heavy applications. Cement kiln dust is produced at kiln inlet, with a temperature around 800°C to 1000°C of the clinker[16–18]. The geopolymer can be produced from any material consists of alumino/silicate reactive materials with few amount of CaO component like metakaoline and strangely alkaline solution. Alternative of Portland cement with its advantage but without its adverse effects ismore environmentally save [19-20]. The produced geopolymer is depending on molar Si:Al ratio of the reacting material generating 3D-polymeric chain ring structure consist of Si-O-Al-O bonds. Geopolymers have been produced through utilizing differentraw materials like: kaolinitic clays [21], metakaolin [22], fly ashes [23-25], blast furnace slag, [26], mixtures of fly ashes and slag [27-28], mixtures of fly ashes and metakaolin [29-30], mixtures of slag and metakaolin[31], mixtures of slag and red mud [32], mixtures of fly ashes and non-calcined materials like kaolin and stilbite [33-35]. All previous materials react with alkaline solutions like: Sodium hydroxide (NaOH) or potassium hydroxide (KOH) or a functional mixture of them. Also, NaOH sodium water glass (Na_2SiO_3) or KOH with potassium water glass (K_2SiO_3) play an important role in the polymerization reaction and decrease the rate of strength development. Egypt has large reserves of kaolin in many areaslike Sinai, Red Sea coast and Aswan, which can meet the needs of local industries for at least 50 years [36]. Kaolin is a lamellar silicate consisting of alternate layers of silica and alumina in tetrahedral and octahedral coordination respectively. Blast furnace slag is also produced in Egypt at a rate of 300 thousand tons/year [37]. Small quantities of these reserves are used as raw materials for road while cement paving and the rest is discharged directly into the lands, which can subsequently pollute the surrounded environment. Therefore, recycling of the environmental hazardous wastes form cement and iron & steel productions will mitigate serious environmental problems and add significant economic benefits to these industries.

The main targets of this work are to evaluate synthesis of eco-friendly geopolymer using some industrial wastes resulting in saving Egyptian resources, added value to these waste and increase investment in cement industry.However; significant progress in financial analysis for geopolymerization process in last 10 years, the cost and the CO_2 abatement potential are not well described [1,26-27].Previous studies on life cycle assessment do not include economic indicators, which is a notable gap to be addressed in further research [38-40].To make the geopolymerization process more effective in direct and indirect reduction of the CO_2 emissions emitted from cement production process, intensive researchesarereally required in the near future. Further evaluations could address integrated assessment ideally using technical, economic, environmental and even social indicators, as well as broadly established methods. Environmental valuations must include direct and indirect environmental impacts and comparisons with other technologies for emissions abatement are necessary. A novel integrated economic and environmental assessment framework is discussed, in which LCA are conducted in parallel and systematically include technology maturity. Then, the goal and the scope of the current study are presented, within the framework of appropriatebenchmark processes and scenarios. Afterward, all the input data and assumptions are described, including the proposed design of the geopolymerization process (section 2). Finally, the analysis and results are presented (section 3).

2. Materialsand Methods

2.1 Materials Resources

The main raw precursors which were used in the experimental work are cement kiln dust (CKD), granulated blast furnace slag (GBFS), silica fume (SF) and metakaolin (MK;the anhydrous calcined form of the kaolin mineral at 650°C for 3h). CKD was supplied from Lafarge Egypt cement company(Suez, Egypt).GBFS and SF were purchased from LOBA chemicals company(Cairo, Egypt).MK was brought from EL-Arieshcement company (Beni-Suef, Egypt).Five kilograms were collected isokinetically from bypass EP-filter screw then homogenized, then particle size analysis using sieve shaker and physical properties were performed as explained in (Table 1). After reaching the required particle size overt mesh 45μm, dried at 105°C for 24h before usage.

2.2 Characterization

The phases composition of the samples were determined using X-ray diffraction (XRD), which carried out by Philips (PW3050/60) diffractometer using a scanning range from 5 to 90 (2Ø), with a scanning speed of 1 Sec./step and resolution of 0.05 degas shown in (Figures (2-5). Surface morphology and microstructure of CKD after drying process was studied by SEM (figure 1), using field emission scanning electron microscope (Gemini (Sigma 500 VP, 2020 version)). The expected major oxides in the CKD, S.F, MK and GBFS were quantified using X-ray fluorescence performed with a comprehensive instrument Panalytical (ARL 9900).CKD is composed of various crystalline peaks affiliated to lime (CaO), calcium hydroxide $Ca(OH)_2$, calcite ($CaCO_3$) and quartz (Figure 2). Albite and microcline are the major minerals detected in XRD-pattern of silica fume material as detailed in (Figure 3). Aluminum oxide and quartz are the main components in Mk as (Figure 4) explains, where while calcium aluminum and calcium silicates are the features of GBFS composites with minor quartz content as seen from (Figure 5).

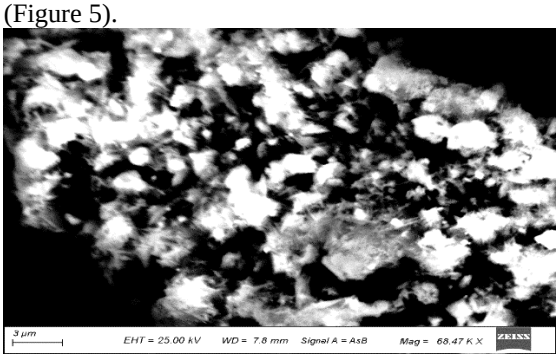


Figure (1):- SEM morphology of CKD microstructure (as received)

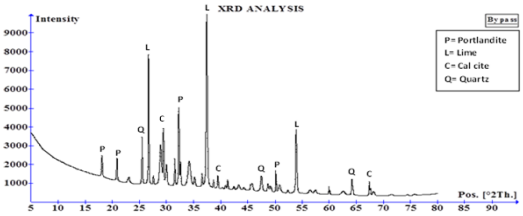


Figure (2):- XRD-patterns of cement kiln dust (CKD).

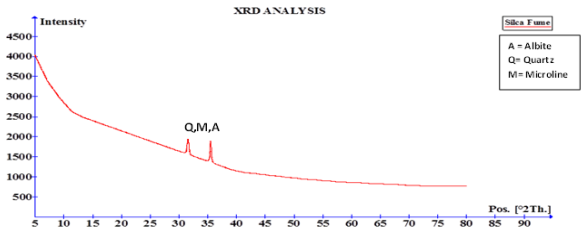


Figure (3):- XRD-patterns of silica Fume (SF).

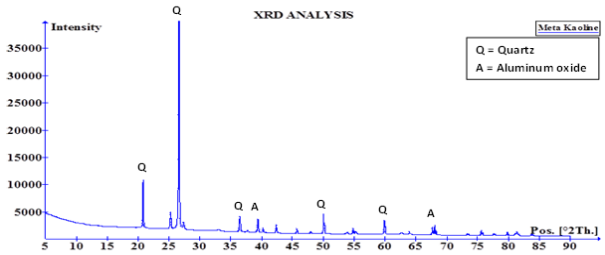


Figure (4):- XRD-patterns of metakaolin (MK).

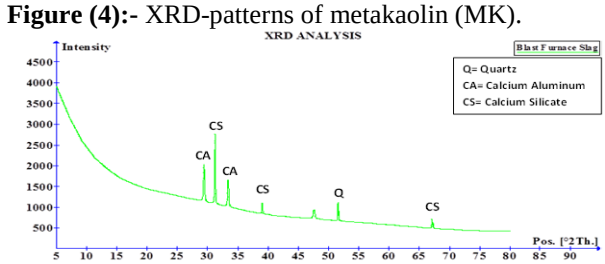


Figure (5):- XRD-patterns of granulated blast furnace slag (GBFS).

Table (1):- Physical Properties of CKD

Characteristic	CKD
X125µm	100.0%
X90µm	85.0%
X63µm	61.0%
X50µm	47.0%
X45µm	33.0%
Plasticity Index	2.22
Specific Gravity Absorption	2.70
Absorption	1.02%
Dry sample real density (kg/dm3)	1.36
Water absorption (wt.%)	0.47

Procedures for absorption and carbonization experiments

Table (2):- Chemical composition of precursors

Material	SiO_2	Al_2O_3	CaO	Fe_2O_3	MgO	SO_3	Na_2O	K_2O	Cl^-
CKD	41.21	32.54	24.68	2.86	0.31	0.41	0.63	0.17	1.46
BFS	32.28	30.56	1.25	0.44	0.06	0.12	0.01	0.01	0.01
SF	94.64	0.97	0.55	0.93	0.35	0.10	0.20	0.25	0.01
MK	67.45	32.69	1.24	0.67	0.05	0.01	0.07	0.06	0.007

The alkaline activator is a mixture of commercial potassium hydroxide and potassiummeta silicate sodium. Potassium hydroxide solution of 8.0M concentration was prepared by mixing 98% pure pallets with distilled water. The mass ratio of SiO2 to K_2O of the potassiummeta silicate solution was 0.7(SiO_2=28.84%, K_2O =0.12% and distilled water =56.6%).The binder mix compositions of the geopolymer pastes were proportioned based on ASTM- C109M,2017 cured at 20°C [41-42], as shown in geopolymerization process (Figure 6).Ten geopolymer paste mixes in two parallel series (MK only, MK blended GBFS, CKD blende silica fume and CKD blended MK) were mixed in the laboratory at 20°C =and R.H95% (Table 3).The alkaline solution content of each mix composition consisted of the same proportion of alkaline solution with a potassium meta silicate to potassium hydroxide ratio of 0.7. The concentration of the KOH solution was 8.0M. The name of mixes was derived from the type of material/s used in its preparation. For instance, MK-CKD-1 represents a geopolymerpaste mix having 50% MK blended CKD. Similarly, the mix designations of S.F-CKD and MK-GBFS represent the MK blended GBFS series respectively. All dry ingredients were first added in a Hobart mixer followed by addition of the activator alkaline solutions to the dry materials and mixed for 3min to 5min. The geopolymer paste specimens of size 140×140×160 mm specimen were cast in accordance with the ASTM-C311,2018 standard [42]. The specimens were remolded after 24h of casting. The specimens were compacted by using a jolting apparatus during the preparation.The paste specimens were cured in room temperature (19±2°C) and at a relative humidity of (95±5%). Compressive strength of the paste specimens was tested at Toni-Technique compressing machine after 7, 28 and 90 days in accordance with the ASTM standard using a loading rate ˷ 0.40MPa/s. A part of the dried yield was kept for SEM and XRD investigations.

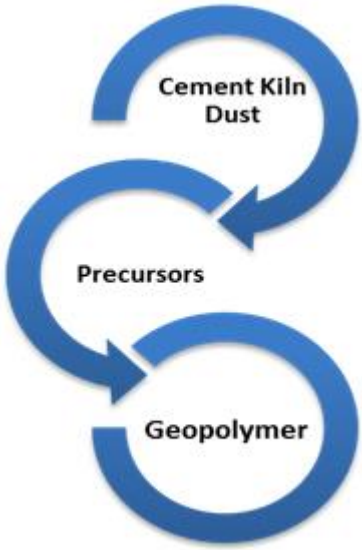


Figure (6):- Geopolymerization process

Table (3):- Mix proportions of geopolymer pastes

Mix	Si/Al	Alkaline Solution (Wt.%)	Mix components (kg/m^3)			
			CKD	MK	GBFS	S.F
CKD	1.26	0.33	500	-	-	-
MK-CKD-0	2.06	0.98	0	500	-	-
MK-CKD-1	1.35	0.51	250	250	-	-
MK-CKD-2	1.35	0.43	300	200	-	-
S.F-CKD-1	1.29	0.44	250	-	-	250
S.F-CKD-2	1.29	0.52	300	-	-	200
GBFS-CKD-1	0.84	0.49	250	250	-	-
GBFS-CKD-2	0.84	0.56	300	200	-	-
MK-GBFS-1	0.62	0.37	-	250	250	-
MK-GBFS-2	0.62	0.37	-	300	200	-

3. Results and Discussions

3.1 Geopolymers Setting time

All geopolymeric mixtures are applicable as they flow easily after mixing (Figure 7). Mixtures can be processed easily enough to pour into molds and pour test samples. Initial and final setting times for geopolymer mixtures were determined by the Vicat instrument. Geopolymers based on MK alone take a long time (> 24h) to set due to their slow rate chemical reaction at room temperature. The surface

charge on the MK particle affects the initial setting properties of the geopolymeric mix during casting[43-44] . On the other hand, geopolymers based on CKD alone take 24h to set due to their cementitious properties when treated at room temperature. The initial setting times of the silica fume geopolymers containing 50% and 66.6% CKD were, 12h and 10h respectively. The final setting times of these mixtures were 21h and 16h respectively. Great reductions in setting times were achieved by blending of the CKD with GBFS. With (1:1) and (3:1) blendingratios, the initial and final setting times of the geopolymer increases from1 to 1.5h and 4.9 to 6.0h respectively. The setting times of the CKD blended GBFS were relatively short compered to the others blends. In addition, setting of MK blended GBFS was also relatively short. Use of (1:1) and (3:1) blending ratios, the initial and final setting times of the geopolymer increases from 1 to 2.7h and 6.0 to 7.5h respectively. Thus, it can be seen that the desired setting time for a wide range application can be reached by controlling the percentage of GGBFS, CKD and SF in the geopolymer mixture.

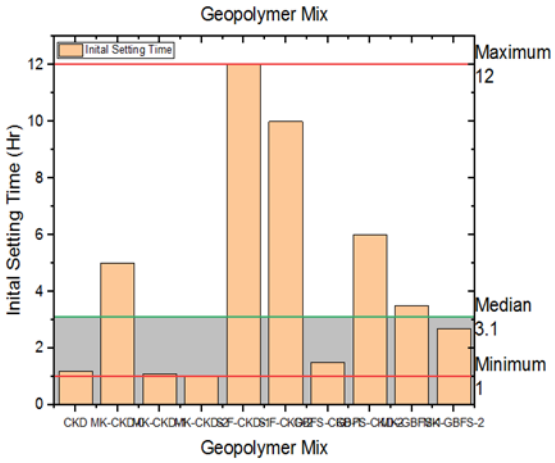
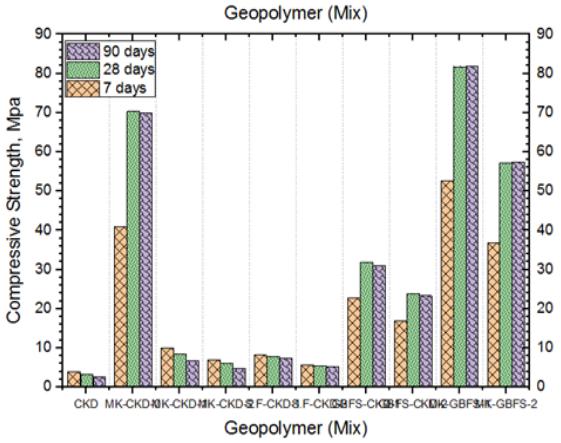


Figure (7):- Initial setting time of geopolymer blends



3.2 Geopolymers compressive strength

The increased rate of geopolymer system development at an stage age can be attributed to the curing heat of reaction treatment and thus measurement of compressive strength (ASTM C39M) was required after 7 days of water curing[41]. Some of the absorbed water inside the paste particles may be evolved during mixing process due to the cracking or damaging of the adhered precursors with saturated water content so, leading to higher workability and durability. It was noted that after 28 days of curing blends can't hardened more and this could be due to absence of the OPC effect during the hydration period. Moreover, (Figure 8) present the compressive strength results of all blends. GBFS pastes reflect high mechanical strength and workability when mixed with MK and CKD respectively than other geopolymer pastes. This could be attributed to the high content of calcium silicate, calcium aluminate and quartz formed the GBFS composites. These results are comparable with those reported by [45] and[46] using conventional silicates. Pastes composed of of GBFS and MK with blending ratio of (1:2) poses low strength by about 69% than GDFS-CKD-1. Replacing MK by CKD mixed with GBFS shows lower strength more than the expected. This is because the CKD has low hydraulic properties, as it contains 58% glass phases (Portlandite + Calcite) and 31% lime content [47-48]. Silica fume pastes have the highest initial setting time and water constancy when mixed with CKD by different portions, this leads to weakness in mechanical and workability properties due to required high curing temperature (60±2°C). Its recommended that producing geopolymer from SF needs specific curing factors due to its high surface area. Visual check of geopolymer specimen shown in (Figure 9).

Figure (8):- Compressive strength of Geopolymer blends

Figure (9):- Visual check of geopolymers specimen CS results.

3.3 XRD and phases clarification

Mineralogical pattern of 50% MK-GBFS-1 geopolymer paste shows in (Figure 9). The area

from 17° to 38° (2θ), known as the important geopolymer characterization regions, wherever they grow, these areas will affect the performance of the compound produced. At peak 21.5 ° and 26.7° (2θ) sharp bands are generated proves the formation of CAH up to 28 days of hydration. This leads to increase the matrix alkalinity as with the progress of thehydration reaction besides the advance of the geopolymerization reaction. The interaction of free alumina with dissolved calcium silicate of the composite forming CAH. With time the C AH fills up and precipitates into empty pores in the later curing age increases the density of the geopolymer and enhance its mechanical properties. A slight increase in calcite may also be observed with an increase in the rate of water due to the carbonation of the atmosphere interactions [51-52]. Hence, (Figure 10) shows the XRD pattern of MK-CKD-0 which has the highest Si/Al ratio that reflect good mechanical properties than other blends as seen in (section 3.2) and proved by XRD. Forming well organized geopolymer leads to improve their mechanical properties. The free $Al^{+}$and calcite tend to form CAH with curing till 28 days, by decreasing the MK % and substituted by CKD on decrease the mechanical strength due to losing of 41% of free Al^{+}. This can be understand from the sharp decrease in the vitreous ands at the above-aforementioned range.Carbonate growth with increased CKD due to reaction under alkaline activation conditions, leads to increase the carbonate content at the blend during the curing times[53].

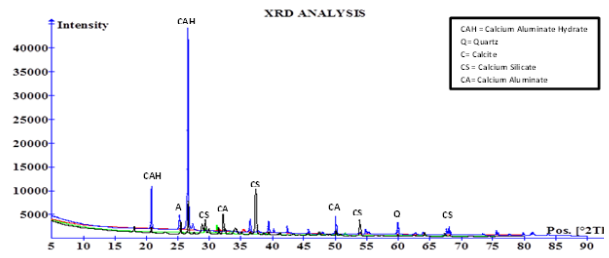


Figure (9):- Mineralogical pattern of 50% MK-GBFS-1 geopolymer paste .

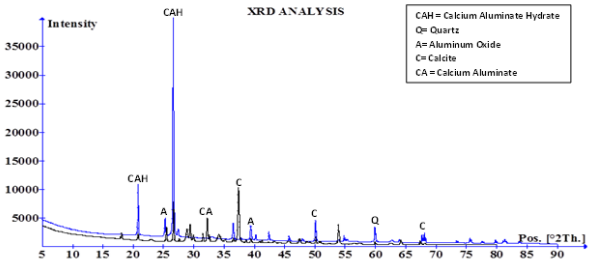


Figure (10):- Mineralogical pattern of MK-CKD-0geopolymer paste .

Conclusion

The main purpose of the current study is to produce environmentally friendly and low cost materials. The influence of cement kiln dust on the setting time, mechanical strength development and workability in different types of geopolymer pastes has been discussed for the room temperature curing condition. CKD was added at rates of 33–50% in the geopolymer pastes based on MK only, MK blended with 33%-50% CKD, silica fume blended with 33%-50% CKD, GBFS blended with 33%-50% CKD and MK blended with 33-50% GGBFS. The strength and phases developments were studied up to the hydration age of 90 days. Compressive strength of room-cured geopolymers was found to increase with the addition of MK at early ages. The 7 -day strength increased by blending of low CKD percent with MK or GGBFS and it increased further with the curing age till three months of hydration. Mechanical strength increased with the addition of MK up to a dosage of 50% and then it declined with further addition .The maximum 90 days compressive strength among all the mixes was 83 MPa which was obtained for the geopolymer paste containing 50% MK and 50% GGBFS. The strengths of this mix were 55MPa and 80MPa at 7 and 28 days respectively .The high Si/Al ratio of the MK-only pastes resulting in good mechanical strength varied between 44MPa and 71MPa at 7 and 28 days respectively. Analysis of the XRD phases showed that adding MK to CKD and GBFS Improving the compressibility of the reacted product. Contribute From MK to form more compact and dense microstructure with better interlocking microstructure leads to enhance the hydraulic properties of geopolymer composites. A safe environment and sustainability of raw materials is the vision of the new century.

Conflict of interest

There is no conflict of interest.

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