**IMPACT: International Journal of Research in Applied,**
**Natural and Social Sciences (IMPACT: IJRANSS)**
**ISSN(E): 2321-8851; ISSN(P): 2347-4580**
**Vol. 2, Issue 5, May 2014, 195-210**

# ECO-FRIENDLY CEMENT FROM CERAMIC WASTE GEOPOLYMARIZATION

**E. EL-FADALY[1], M. R. MOSTAFA[2], M. E. I. SARAYA[3], F. A. NASSAR[4], T. EL-SOKKARY[5]**
**& H. EL-DIDAMONY[6]**

[1]Environmental Studies and Research Institute, Sadat City University, Sadat City, Egypt

[2,3,4]Faculty of Science, Al-Azhar University, Cairo, Egypt

[5]Housing and Building National Research Center, Cairo, Egypt

[6]Faculty of Science, Zagazig University, Zagazig, Egypt

## ABSTRACT

In ceramic tile production, because of various reasons, unsold fired products come out. These are waste tiles and only a little part of them is reused. If these waste tiles were used in geopolymer production, this pollution decreases. In this study, usage of waste tile as pozzolan was studied. The effect of sodium silicate solution with different molar ratio on the compressive strength of Egyptian ceramic waste -based geopolymers is investigated. Three different alkaline solutions with different ratio (10-40%), with different soluble silica contents were used to activate ceramic waste. The primary reaction product was a sodium aluminosilicate gel, while different types of zeolites appeared as minority phases. The percentage and composition of these reaction products were found to depend on both the soluble silica content present in the activating solutions and curing time. In addition, the amount of gel was observed to have a decisive effect on the mechanical strength developing in the material. X-ray diffract grams of geopolymers indicated the existence of the major amorphous phases of ceramic waste, as well as the formation of a new amorphous phase in the geopolymeric matrices. Fourier-transform infrared (FTIR) spectroscopy analysis revealed essential ceramic waste phase transformations within geopolymers that affected their mechanical strength. The results of the current research indicate that a compressive strength is increased with the increase of alkali content, as well as with the increase of sodium silicate in the synthesis of geopolymers. These results enhanced usage of alkali activated ceramic tile waste as eco-friendly cement in adhesion and building materials without more emission of $CO_2$.

**KEYWORDS:** Alkali Activation, Ceramic Waste, Compressive Strength, DTA, XRD, FT-IR

## INTRODUCTION

Geopolymers are chemically hardened monolithic aluminosilicate gels formed by partial dissolution and polycondensation of aluminosilicate materials, such as fly-ash and calcined clays, in aqueous alkaline environments [1, 2]. Geopolymers have been proposed as an alternative to traditional Ordinary Portland Cement (OPC) for use in construction applications, due to their excellent mechanical properties [3], low temperatures required for synthesis and their intrinsic fire resistance [4]. Like zeolites and some aluminosilicate gels, geopolymers are synthesised in aqueous media, albeit much lower water weight fractions, typically less than 35%. Due to their excellent mechanical properties, the bulk of literature is focussed on the effect and application of different raw materials on the compressive strength of geopolymers [5], chemical impurities [6], and the effect of the chemical composition of the alkali activating solutions [7]. Only a relatively small number of investigations have specifically studied the effects of high-temperature on geopolymeric gels [8–13].

Studies of geopolymeric gel have not explored the effect of gel Si/Al, alkali cation, raw material or impurities. Each of these factors plays crucial roles in determining the mechanical properties of geopolymers at ambient conditions and should also be expected to affect greatly the properties of geopolymers at elevated temperatures. The geopolymer structure is formed from tetrahedrally co-ordinated aluminium and silicon atoms bridged with oxygen [14, 15]. The negatively charged aluminium in IV-fold coordination is balanced via association with monovalent cations, typically sodium and potassium, provided by the alkaline activating solution [1]. The resulting inorganic gel is amorphous to electron diffraction [1, 9, 6], and exhibits a single broad hump in $^{29}$Si MAS-NMR spectra, similar to aluminosilicate gels [15, 16]. However, transformation of the amorphous gel into zeolite has been observed, where structural reorganisation is rapid due to an open microstructure [5, 17, 18]. The microstructure of geopolymers has been observed to vary greatly depending on the Si/Al, a result of variation in the density of the aluminosilicate gel.

The change in microstructure appears to be very important to the mechanical properties of geopolymers, with strength observed to increase generally with decreasing gel density [17]. Rahier et al. [13] broadly investigated some thermal properties of a geopolymer of non-prescribed composition synthesised from metakaolin and sodium silicate solution (denoted Na-geopolymer), primarily the dehydration and thermal shrinkage. The specimen investigated by Rahier exhibited shrinkage of approximately 6% during dehydration, without significant densification observed at higher temperatures (>600 $^{o}$C). More recently, the thermal characteristics of Na-geopolymer with a Si/Al of approximately two has been investigated, revealing a similar region of shrinkage associated with dehydration, but also the specimen was observed to densify at approximately 800 $^{o}$C [8]. The different thermal shrinkage behaviour of these systems suggests that there are differences in the thermal behaviour of Na-geopolymers with differing composition which is yet to be elucidated by systematic investigation. Furthermore, the dehydration and densification processes occurring in Na-geopolymers have not been determined. The microstructure of Na-geopolymer is known to be highly dependent on Si/Al, which may play a significant role in determining the material’s thermal properties and the effect of elevated temperatures on its physical properties [17].

These two elements are able to form highly concentrated aqueous solutions and solvate large amounts of silicon and aluminium, both of which are critical for geopolymerisation. Geopolymeric materials synthesised with different alkali cations exhibit marked differences in Si/Al ordering [14,15], thermal stability [8,6] and setting properties [6]. To some extent, the effect of alkali cation on the mechanical properties of geopolymers has been observed [6, 19-22]. Despite the apparent effects of alkali, there has been no systematic investigation to determine the extent to which alkali type will affect the development of the mechanical properties of geopolymers. Alkali cations are associated with aluminium, where the AlO4 groups have a single negative charge due to aluminium (III) being in tetrahedral co-ordination. A study of the effect of alkali cations (sodium and potassium) on the molecular ordering of the geopolymer gel concluded that the concentration of silicon in the activating solution plays a large role in aiding the incorporation of aluminium into the matrix via solution phase speciation with silicon. Similarly, the nature of the alkali cation also contributes to the level of reaction of the solid aluminosilicate source, as well as the level of aluminium incorporation [14]. The effect of the concentration of silicon in the alkali activating solution has been investigated on systems utilising both fly-ash [22, 23] and metakaolin [18, 24] as solid aluminosilicate sources. Furthermore, the relationship between geopolymer composition, microstructure and mechanical properties has recently been conducted by the study of geopolymers synthesised from metakaolin with composition $Na(SiO_2)$ zAlO2•5.5$H_2O$, where $1.15 \le z \le 2.15$ [24].

Geopolymers possess excellent physico-chemical and mechanical properties, including low density, micro-porosity or nano-porosity, negligible shrinkage, high strength, thermal stability, high surface hardness, fire and chemical resistance [25, 26, 10]. Due to these properties, they are viewed as alternative materials for certain industrial applications in the areas of construction, transportation, road building, aerospace, mining and metallurgy. Certain industrial wastes, such as coal-fired fly ash, blast furnace slag and mine tailings contain sufficient amounts of reactive alumina and silica, in order to be used as source materials for the geopolymerization process. Therefore, geopolymerization can be considered as an economically viable technology for the transformation of industrial wastes and/or by-products of aluminosilicate composition into attractive construction materials [25, 27, 28]. The aim of the present study was to explore the effect of the sodium oxide and soluble silica content in a series of activating solutions on the physico-mechanical and microstructural properties of ceramic waste – base geopolymer to identify and quantify such products and their impact on mechanical development in the material.

## MATERIALS AND METHODS

The materials used in this investigation were ceramic waste, Commercial grade sodium hydroxide in pallets form (97% -100% purity) as well as sodium silicate solution ($Na_2O$=10.3%, $SiO_2$=28.9%, Water = 60.8%) were used to preparation of alkali activators. The ceramic waste was activated with a series of alkaline solutions, all with a practically constant silicon oxide content (≈28%), but with varying proportions of sodium oxide. The chemical composition and certain other properties of each solution are given in Table 1 and molar oxides ratio used in the production of geopolymers from ceramic tile waste are given in Table 2, which also shows the differences in both the sodium oxide and the water content achieved by mixing sodium silicate and sodium hydroxide in different proportions. Three different alkali solutions (AA1-AA3) with different soluble silica contents were used as ratio 10-40%, to activate ceramic waste.

**Table 1: Chemical Analysis of Starting Materials, Mass %**

| Oxide Content % | Ceramic Tile Waste | Sodium Silicate | AA1 | AA2 | AA3 |
|---|---|---|---|---|---|
| $SiO_2$ | 47.23 | 30.7 | 30.7 | 30.7 | 30.7 |
| $Al_2O_3$ | 17.39 | -- | -- | -- | -- |
| $Na_2O$ | 10.08 | 10.3 | 14.3 | 18.3 | 29.3 |
| CaO | 1.06 | -- | -- | -- | -- |
| MgO | 14.77 | -- | -- | -- | -- |
| $K_2O$ | 3.70 | -- | -- | -- | -- |
| $SO_3$ | 0.15 | -- | -- | -- | -- |
| $Fe_2O_3$ | 0.01 | -- | -- | -- | -- |
| $TiO_2$ | 0.01 | -- | -- | -- | -- |
| $P_2O_5$ | 4.76 | -- | -- | -- | -- |
| $H_2O$ | 0.00 | 59.0 | 55.0 | 51.0 | 40.0 |
| L.O.I | 0.79 | -- | -- | -- | -- |
| **Total** | **99.21** | **100.0** | **100.0** | **100.0** | **100.0** |

**Table 2: Molar Oxide Ratios Used in the Production of Geopolymers from Ceramic Tile Waste**

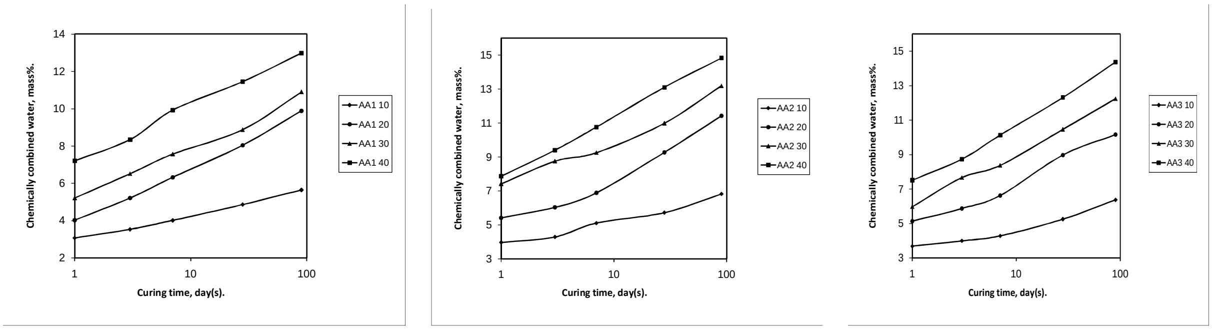
| AA1 | $SiO_2$: $Al_2O_3$ | $Na_2O$: $SiO_2$ | $H_2O$: $Na_2O$ | AA2 | $SiO_2$: $Al_2O_3$ | $Na_2O$: $SiO_2$ | $H_2O$: $Na_2O$ | AA3 | $SiO_2$: $Al_2O_3$ | $Na_2O$: $SiO_2$ | $H_2O$: $Na_2O$ |
|---|---|---|---|---|---|---|---|---|---|---|---|
| **10** | 2.89 | 0.22 | 1.76 | **10** | 2.89 | 0.23 | 1.58 | **10** | 2.89 | 0.26 | 1.36 |
| **20** | 3.07 | 0.24 | 1.83 | **20** | 3.07 | 0.25 | 1.65 | **20** | 3.07 | 0.29 | 1.29 |
| **30** | 3.24 | 0.25 | 1.86 | **30** | 3.24 | 0.27 | 1.63 | **30** | 3.24 | 0.33 | 1.17 |
| **40** | 3.42 | 0.26 | 1.73 | **40** | 3.42 | 0.29 | 1.47 | **40** | 3.42 | 0.36 | 0.97 |

Mixing of all the materials were done manually in the laboratory at room temperature. The ceramic waste and the alkali solution were mixed homogeneously. The “solution/ash” ratio used was 0.5 by weight. The fresh geopolymer paste was used to cast cubes of size 2x2x2 cm to determine its compressive strength. Water content of the sodium silicate mixture was adequate to facilitate geopolymerization without deterioration of the mechanical properties of the geopolymer. The specimens were prepared according to the method followed by Hardjito et. al. [28]. Each cube specimen was cast in three layers by compacting manually as well as by using vibrating machine. The specimens were wrapped by plastic sheet to prevent loss of moisture and placed in an oven. The bulk density was determined from weights of hydrated paste in air and suspended in water according to Archimedes principle [29]. The determination of compressive strength of cement pastes was carried out according to ASTM designation: C 109-80 [30] using compressive strength machine. After each experiment, the material was grinded (sizeb65 μ) and mixed with a small volume of acetone in order to dehydrate the system and then to prevent the material evolution. The combined water (Wn), was determined from the ignition loss of dried samples at 1000 °C. The XRD pattern ceramic waste -base geopolymer was recorded on (M/S. Shimadzu Instruments, Japan) diffractometer XRD 7000 with Ni filtered CuKα as a radiation source at 2θ scan speed of $4^{\circ}$ $\text{min}^{-1}$. XRD. IR spectrometer (Perkin-Elmer 880). The FTIR spectra in the wave number range from 400 to 4000 $\text{cm}^{-1}$ were obtained in atmosphere using the KBr pellet technique. The pellets were prepared by pressing a mixture of sample and dried KBr (sample : KBr approximately 1:200) at 8 $\text{t/cm}^2$. Finally, for the characterization of the surface morphology and the analysis of microstructure compositions a JOEL JSM-type scanning electron microscope was used.

## RESULTS AND DISCUSSIONS

### Chemically Combined Water Contents

The chemically combined water contents of alkali activated ceramic waste cured at room temperature with different alkali activators up to 90 days, are seen in Figures (1, a-c). It is clear that the chemically combined water contents increase with time for all activated pastes. This is mainly due to the progress of alkali reactions of activator with ceramic waste phases. As the activation time of granulated slag progresses, more hydration products are formed then the chemically combined water content increases. Also the chemical combined water content increases with sodium hydroxide from AA1 into AA2 and then decreases at AA3. This is mainly due to the progress of alkali activation reaction and formation of hydration products that have water high contents. This is also due to the decrease of $Na_2O/SiO_2$ which tends to form more CSH with higher water contents [31].



(a) (b) (c)

Figure 1: Chemically Combined Water Contents of (a) Alkali Activated Ceramic Waste by (a) AA1 (b) AA2 and (c) AA3, Cured at Room Temperature, up to 90 Days

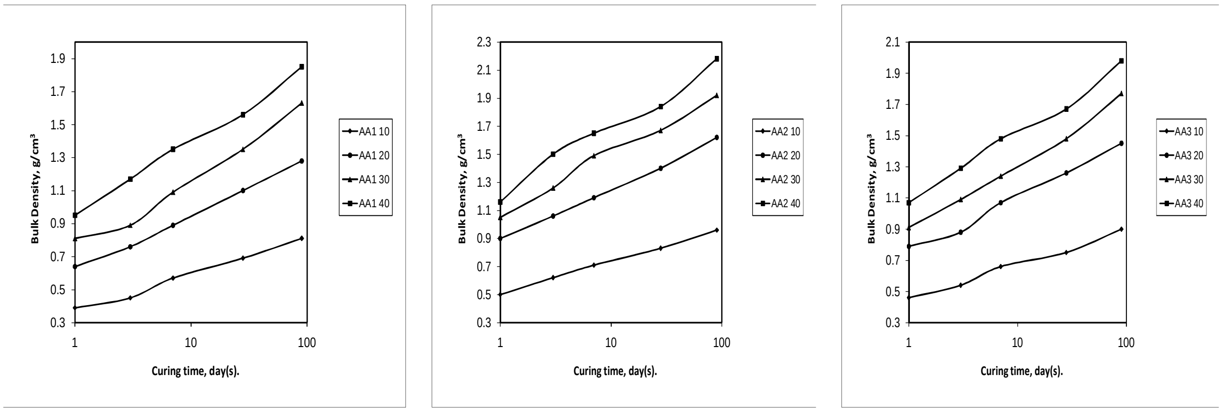
## Bulk Density

The bulk density of alkali activated ceramic waste cured at room temperature up to 90 is seen in Figures (2a-c). It is clear that the bulk density increases with curing time up to 90 days, for all activated pastes as a result of the activation reaction of alkali activator with ceramic waste components. On the other side the bulk density increases with alkali content from AA1 into AA2 and then decreases at AA3. This is mainly due to increasing the hydration products. This hydrate have tendency to leading then the porosity increases, therefore, the bulk density decreases. The bulk density values of the activated ceramic waste are lower than those of the alkali activated slag. This may be due to the decrease of $Ca^{2+}$ in the ceramic waste; therefore, the activated ceramic wastes have lower amounts of CSH which is the main source of mechanical properties of the cement pastes. As the $Na_2O$ content increases the bulk density decreases due to the formation Na. rich CSH with low bulk density and high porosity [32, 33].

## Compressive Strength

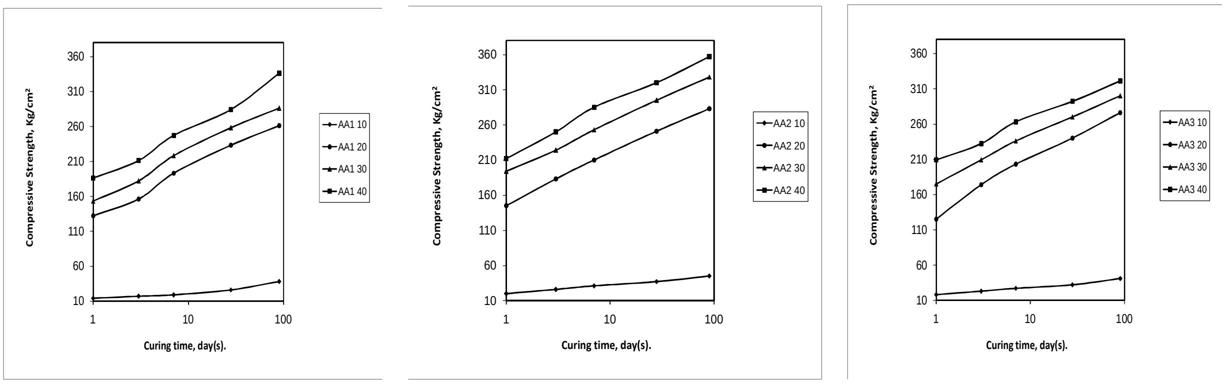
The compressive strength values of alkali activated ceramic waste cured at room temperature with different types of alkali activators up to 90 days, are seen in Figures (3 a-c). It is clear that the compressive strength of activated ceramic waste increases with time for all activated pastes. Also, the compressive strength increases with the temperature as well as alkali content from AA1 up to AA2 and then decreases at AA3. This is mainly due to the increasing of dissolution of component with alkali and enhances the performance of geopolymerization reaction. Also, the compressive strength increases with the$SiO_2/Al_2O_3$ due to the increases of CSH as the main factor of compressive strength.

As a result of the free alkali formation and dilution effect in addition to the formation of Na-Calcium aluminosilicate hydrate with low strength. The low Na-CASH and CSH gives low strength. Formation of the geopolymer framework is greatly dependent up on the dissolution of the ceramic waste particulates, which is directly affected by concentration of alkali solution. Activator solution with higher concentration of NaOH up to AA2 provides better dissolving ability to ceramic waste particulates and produces more reactive bond for the monomer, which will increase inter-molecular bonding strength of the geopolymer. The compressive strength of alkali activated ceramic waste is lower than those of the alkali activated slag. This may be due to the decrease of $Ca^{2+}$ in the ceramic waste; therefore, the activated ceramic wastes have lower amounts of CSH which is the main source of mechanical properties of the cement pastes [33].



(a) (b) (c)

**Figure 2: Bulk Density of Alkali Activated Ceramic Waste by (a) AA1, (b) AA2 and (C) AA3 Cured at Room Temperature, up to 90 Days**



**(a)** **(b)** **(c)**

**Figure 3: Compressive Strength of Alkali Activated Ceramic Waste by (a) AA1, (b) AA2 and (C) AA3, Cured at Room Temperature, up to 90 Days**

## FTIR Spectra

Figure (4) shows FTIR-Spectra of alkali activated ceramic waste cured at room temperature at 90 days, with different types of alkali activators. It is seen in Figure 4, that the band centered at about 1016 cm-1 in raw ceramic waste, corresponding to the Si–O asymmetric stretching in tetrahedral [34, 35] is shifted to lower wavenumbers. That increases with alkali concentration up to A5 and then return to higher wavenumbers. Also, the intensity of band increases with alkali concentration.

The shift toward the low wave number may be attributed to the partial replacement of $SiO_4$ tetrahedron by $AlO_4$ tetrahedron, resulting in a change in the local chemical environment of Si–O bond. The formation of the geopolymer is also indicated by the band at about 870 cm1 linked to Al–O symmetric stretching in tetrahedral [35], which is absent in the spectrum of ceramic waste.

Other bands related to the formation of the geopolymer are those at about 1420 and 1650 cm-1 related to O–H bending and H2O stretching, respectively [36]. The band at about 1460 cm-1 is related to the formation of carbonate by reaction of alkali metal hydroxide with atmospheric $CO_2$.(A1=10%AA1, A2=20%AA1, A3=30%AA1, A4=30%AA2, A5=40%AA2, A6=20%AA3, A7=30%AA3 and A8=40%AA3)

Figure (5) shows FTIR-Spectra of alkali activated ceramic waste with A5 (40%AA2) activator, cured at room temperature up to 90 days. It is shown that the intensities of the bands related to the geopolymer formation increase as the polycondensation time increases. On the other side, the intensity of characteristics band of slag decreases with time. This is mainly due to the dissolution of ceramic waste components in the alkali medium and shafted to lower wavenumbers.

The sequence of spectra in this figure also shows that the band centered at about 460 cm1 (Si–O bending) undergoes a very small shift towards higher wave numbers as a consequence of the incorporation of alumina in the geopolymer [35]. This band would be shifted much more considerably towards higher wavenumbers in the case of a geopolymer only made of SiO4 tetrahedral [37]

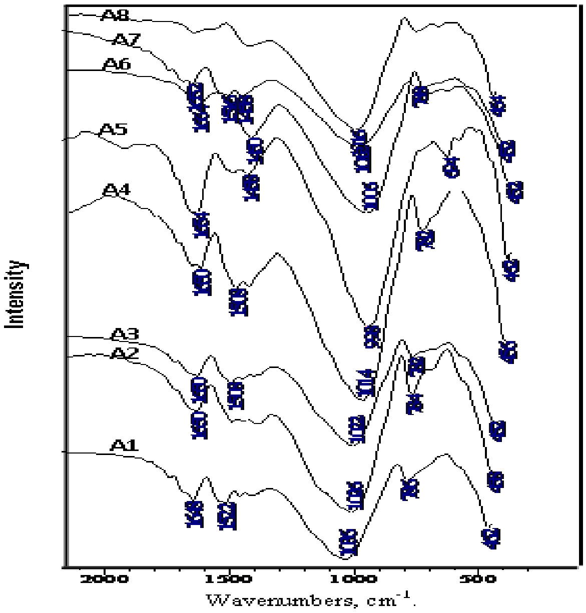


Figure 4: FTIR Spectra of Alkali Activated Ceramic Waste with Different Types of Activator, Cured at 90 Days

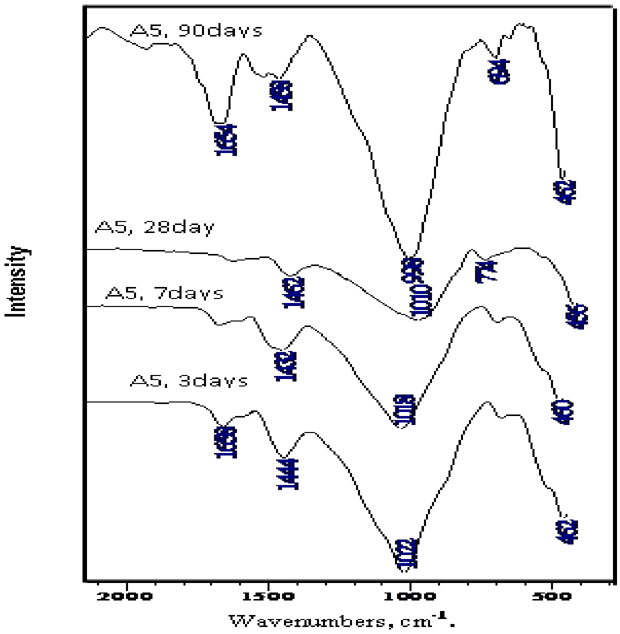


Figure 5: FTIR Spectra of Alkali Activated Ceramic Waste with Activator A5, Cured up to 90 Days

## XRD Pattern

Figure (6) show the pattern of XRD of ceramic waste, where it is a mixture of different crystals and amorphous phase. Figure (7) show XRD pattern of alkali activated ceramic waste cured at room temperature at 90 days, with different types of alkali activators. Ceramic waste based geopolymer have a large diffuse halo peak at about 20–40◦ (2θ max Cu K). This means that the ceramic waste based geopolymeric products are mainly X-ray amorphous materials.

Also, indicating the formation of an alkaline aluminosilicate gel [38]. In addition, several sharp characteristic peaks are also seen. The other signals detected corresponded to zeolite structures such as hydroxylsodalite

(Na4Al3Si6O12-OH, JCPDS 11–0401) and herschelite (NaAlSi2O63H2O, JCPDS 19–1178) or alkaline bicarbonates. Other zeolite-type crystalline phases also appeared after activation, which varied depending on the nature of the activating solution used and curing time [39].

Figure (8) shows XRD pattern of alkali activated ceramic waste with A5 activator, cured at room temperature up to 90 days. The intensity of the characteristic peaks of zeolite products decreases with time and also appears new at other position. These changes can be explained by the fact that zeolite are metastable and may undergo successive transformation into one or several more stable phases [39].

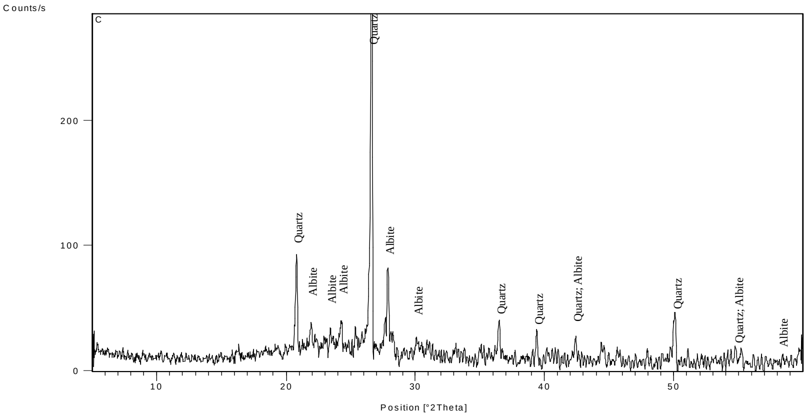


Figure 6: XRD Pattern of Ceramic Tile Waste

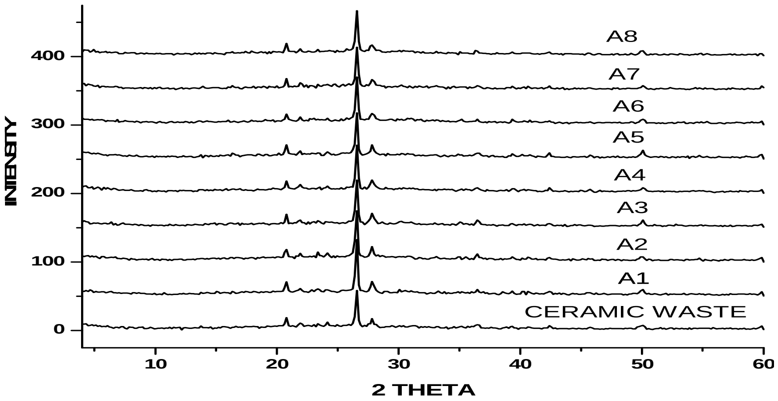


Figure 7: XRD Pattern of Alkali Activated Ceramic Waste with Different Activators, Cured at 90 Day

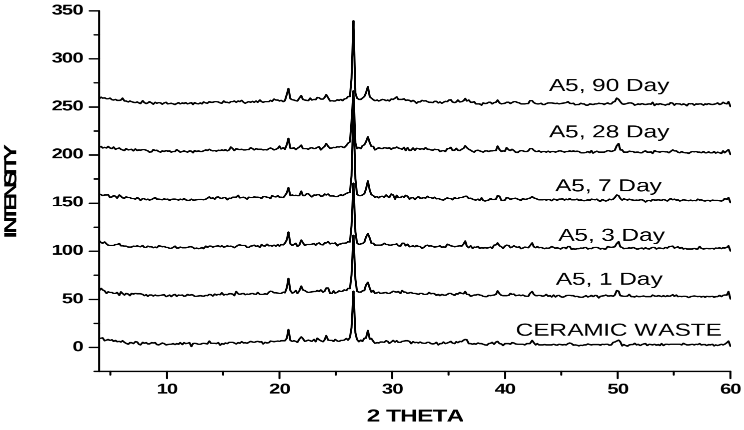


Figure 8: XRD Pattern of Alkali Activated Ceramic Waste with Different Activators, Cured up to 90 Day

## Thermal Analysis

DTA curves of the geopolymer pastes (A4, A5, A6) cured at room temperature up to 90 days is reported in Figure (9). These show some endothermic peaks around 100-250°C, 450 and 800ºC. According to the literature, the final products of geopolymerization contain physically bonded, chemically bonded water and hydroxyl groups (OH) [40]. Independently of the composition, endothermic peaks, observed between 100 and 250 ºC, should be ascribed to the phenomena of dehydration and dehydroxylation of free water and bound water [41]. Two small peaks were also found at 450 and 800ºC indicating the decomposition of $Ca(OH)_2$ and carbonates formed as a result of the incorporation of alkaline solution and high calcium content in slag [42]. On the other side, there is a small densification which continues up to 900°C, indicating a good thermal stability of all the formulations tested [43-44]. The cement A5 shows the higher endothermc peak in the range up to 250°C indicating the maximum activation of ceramic wastes.

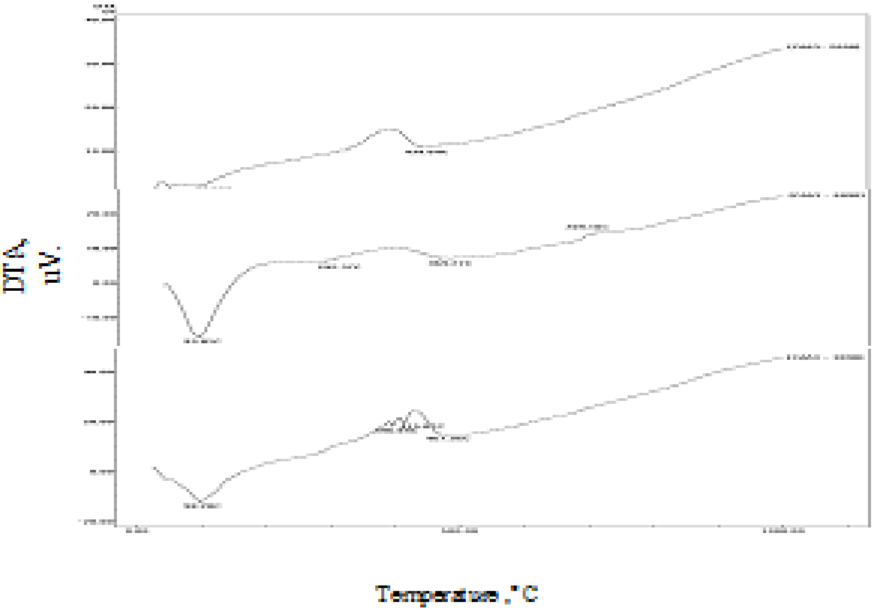


Figure 9: DTA Curves of Alkali Activated Ceramic Waste with Different Types of Activator (A4, A5 and A6 from the Top)

## Scan Electron Microscope Investigation

The scanning electron micrograph of ceramic waste based geopolymer activated wit A5 cured at ambient temperature up to 90 day are shown in Figures (10,11). This sample gave the highest value of compressive strength. Microstructural observations by SEM were very much consistent with the phase transformations observed by XRD. The microstructure of mixtures was characterized by dense and homogeneous phases which accounted for their high strengths. However, a detailed examination (at higher magnification) of the amorphous-looking phases revealed that, there were embedded pockets of crystalline growths [35, 45]. Irregularly shaped macropores are clearly seen as well as spherical cavities which either can be due to air bubble entrapment or are part of the interface between the unreacted spherical ash particles and the geopolymeric matrix. Simplified geopolymerization reactions can be described as initial dissolution of the aluminium and silicon from ceramic waste followed by their polymerisation, gelation and hardening. Van Deventer and co-workers suggested that presence of calcium in solid waste materials will provide extra nucleation sites for precipitation of dissolved species and cause rapid hardening [46].

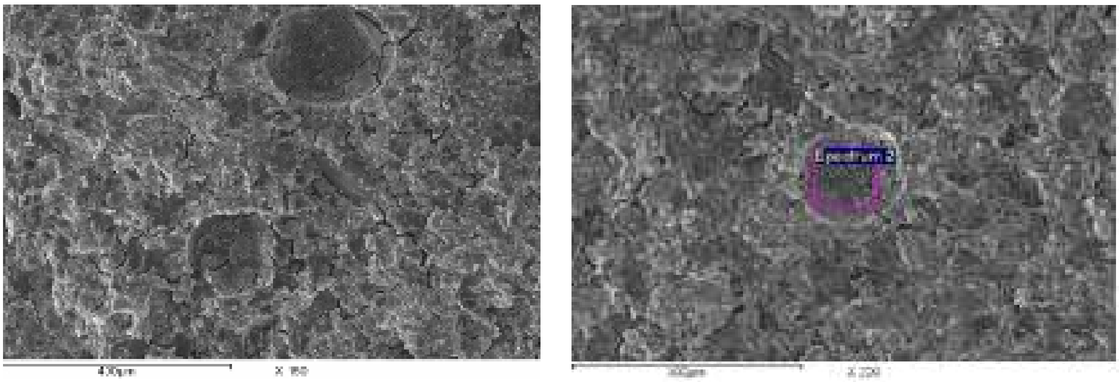
According to the classification scheme proposed by Davidovits, when Si:Al=1 it is a sialate (–Si–O–Al–O–), for Si:Al=2 it is a sialatesiloxo (–Si–O–Si–Al–O) for Si:Al=3 it is a sialate-disiloxo (–Si–O–Al–O–Si–O–Si–) and when Si:Al>3 it is deemed to be sialate link, poly (sialate-multisiloxo) [1]. Generally, the three-dimensional amorphous geopolymeric network with a general formula of Rn–[[–Si–O2]z–Al–O]n w$H_2O$ (R represents a cation such as sodium, potassium, or calcium; n is the degree of polycondensation; z = 1, 2, or 3; and w is the amount of binding water) is often argued to be the phase that gives the binding property to the geopolymeric gel. The ratio of Si/Al should be 1–3, and the ratio of Na/Al should be 1. During the geopolymerization reaction process, Si/Al ratios in the original ceramic waste, the reactive ceramic waste sphere, and the reaction products of ceramic waste are changing [47].

The physical and mechanical properties of geopolymers were a function of the $SiO_2/Al_2O_3$ and $Na_2O/Al_2O_3$ ratios [48]. EDXA analysis is conducted on the sample and the results are listed in the Tables (3,4). It was found that the elemental composition was dominated by Si, Al and Na as well as Ca. According to the chemical compositions and The $SiO_2/Al_2O_3$ ratios of 4.5 and 3.6, and $Na_2O/Al_2O_3$ ratios of 1.90 and 0.34 confirmed the presence of continuous mass of strong geopolymer. Also, it is suggested that phase might be a geopolymeric binder, which has similar characteristics to the PSS type of geopolymeric binder. This observation is also consistent with previous observations by Yip et al. [49].

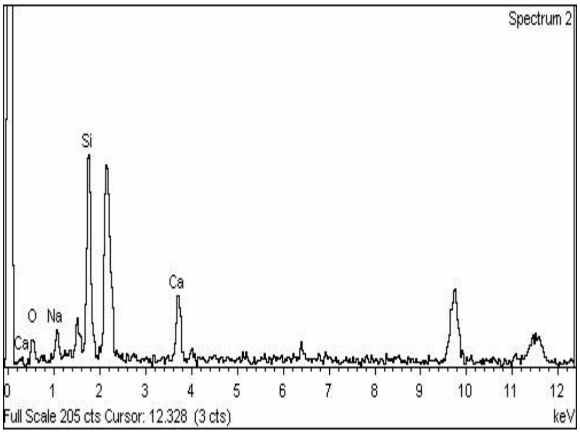
## CONCLUSIONS

The need to reduce the consumption of energy and the release of CO2 is recognized in the Portland cement industry and much effort or resources have been expanded in improving the traditional manufacturing processes. So, there are important needs to resolve the ecological and economic problems and in the same time solving ordinary Portland cement problems like sulfate, chloride and sea water attack. Recycle of solid wastes as ceramic tile waste, can obtain binder materials which characterized by resistance to aggressive media, cheaper and friend to environment than Portland cement. The activation of ceramic tile waste can be occurred by different activators for dissolution the network of silicates and aluminates as well as calcium ion which react with each other to form geopolymers and hydrated products. In this work mixture sodium hydroxide and sodium silicate liquid with different proportions are used as activators. The hydraulic properties of different pastes are followed by the determination of combined water content, bulk density and compressive strength. The hydration products were also identified by IR spectroscopic technique, and SEM observation of some selected pastes. The following results are concluded: ceramic tile waste activated by different activators (AA1, AA2 and AA3) and different proportions (10%, 20%, 30% and 40%) were mixed with water and cured in humidity up to 90 days. The combined water contents increases with curing time up to 90 days. The increase of liquid sodium silicate proportion increases the combined water contents from AA1 until AA2 and low decreases with AA3.

The bulk density increases with curing time up to 90 days. The increase of liquid sodium silicate proportion increases the bulk density from AA1 until AA2 and low decreases with AA3. The compressive strength increases with curing time up to 90 days. The increase of liquid sodium silicate proportion increases the compressive strength from AA1 until AA2 and low decreases with AA3. The increase of liquid sodium silicate proportion and temperature increases the physico-mechanical properties from AA1 until AA2 and low decreases with AA3. The best alkali-activators which higher physico-mechanical properties which mixed with water and cured in humidity up to 90 days was AA2.



(a) Low Magnification Micrograph (b) High Magnification Micrograph

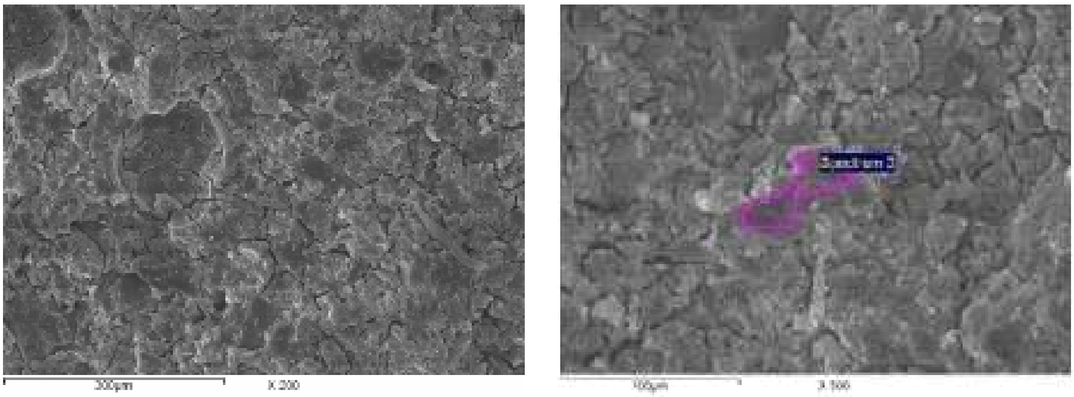


(c) EDAS Analysis and Chemical Composition

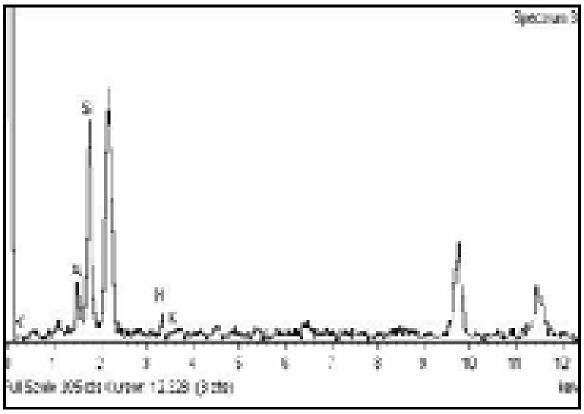
Figure 10: SEM Images and EDAS Spectra of Ceramic Waste geopolymeric Paste with A5 Cured at Room Temperature, up to 90 Days

Table 3: EDXA Analysis is Conducted on the Sample

| Element | App Conc. | Intensity Corrn. | Weight% | Weight% Sigma | Atomic% |
|---|---|---|---|---|---|
| O K | 0.06 | 0.4271 | 41.99 | 7.41 | 56.37 |
| Na K | 0.02 | 0.5053 | 12.33 | 3.55 | 11.52 |
| Si K | 0.08 | 0.6830 | 33.31 | 4.72 | 25.48 |
| Ca K | 0.04 | 0.8853 | 12.37 | 2.10 | 6.63 |
| **Totals** | | | **100.00** | | |



(a) Low Magnification Micrograph (b) High Magnification Micrograph



(c)

**Figure 11: Other SEM Images and EDAS Spectra of Ceramic Waste Geopolymeric Paste with A5 Cured at Room Temperature, up to 90 Days**

**Table 4: EDXA Analysis is Conducted on the Sample**

| Processing : Element | App | Intensity | Weight% | Weight% | Atomic% |
|---|---|---|---|---|---|
| | **Conc.** | **Corrn.** | | **Sigma** | |
| Mg K | 0.02 | 0.5495 | 7.07 | 1.78 | 9.37 |
| Al K | 0.03 | 0.6265 | 9.78 | 1.66 | 11.67 |
| Si K | 0.11 | 0.6508 | 35.49 | 2.14 | 40.68 |
| Ca K | 0.20 | 0.9200 | 47.66 | 2.13 | 38.28 |
| **Totals** | | | **100.00** | | |

## REFERENCES

1. Davidovits. J., “Geopolymers: Inorganic polymeric new materials”. (1991) J. Therm Anal 37(8) 1633.
2. Palomo A., Grutzeck M.W., Blanco M.T., (1999) “Alkali-activated fly ashes: cement for the future” Cement Concrete Res 29(8) 1323.
3. Rowles M., O’Connor. B. (2003) “Chemical optimization of the compressive strength of aluminosilicate geopolymers synthesized by sodium silicate activation of metakaolinite” J Mater Chem. 13(5) 1161.
4. Davidovits N., Davidovics M., Davidovits J., (1989) “Ceramic–ceramic composite material and production method”. USP 4,888, 311.
5. Yip, C. K., Lukey, G. C. and van Deventer, J. S. J. (2003) "Effect of blast furnace slag addition on microstructure and properties of metakaolinite geopolymeric materials." Ceramic Transactions 153: 187-209.
6. Van Jaarsveld, J. G. S. and van Deventer, J. S. J. (1999) "Effect of the alkali metal activator on the properties of fly ash-based geopolymers." Industrial & Engineering Chemistry Research 38(10): 3932-3941.
7. Phair, J.W. & Van Deventer, J.S.J., (2002) “Effect of silicate activator pH on the microstructural characteristics of waste-based geopolymers”, International Journal of Mineral Processing, vol. 66, nos. 1-4, pp. 121-143.
8. Barbosa V.F.F. and MacKenzie. K.J.D. (2003) Thermal Behavior of Inorganic Geopolymers and Composites Derived from Sodium Polysialate. Mater. Res. Bull., 38, 319-31, (2003).

9. Barbosa V.F.F. and MacKenzie K.J.D. (2003) “Synthesis and Thermal behavior of Potassium Sialate Geopolymers”. Mater. Lett., 57, 1477-82.
10. Cheng T.W., Chiu J.P., (2003). “Fire-resistant geopolymer produced by granulated blast furnace slag” Mine Eng. 16(3) 205.
11. Rahier, H., Simons, W., van Mele, B. and Biesemans, M. (1997). "Low-temperature synthesized aluminosilicate glasses. 3. Influence of the composition of the silicate solution on production, structure and properties." Journal of Materials Science 32(9): 2237-2247
12. Rahier, H., van Mele, B., Biesemans, M., Wastiels, J. and Wu, X. (1996.b) "Low-temperature synthesized aluminosilicate glasses. 1. Low-temperature reaction stoichiometry and structure of a model compound." Journal of Materials Science 31(1): 71-79.
13. Rahier, H., van Mele, B. and Wastiels, J. (1996) "Low-temperature synthesized aluminosilicate glasses. 2. Rheological transformations during low-temperature cure and high-temperature properties of a model compound." Journal of Materials Science 31(1): 80-85.
14. Duxson, P., Lukey, G. C., Separovic, F. and van Deventer, J. S. J. (2005.b) "The effect of alkali cations on aluminum incorporation in geopolymeric gels." Industrial & Engineering Chemistry Research 44(4): 832-839.
15. Duxson, P., Provis, J. L., Lukey, G. C., Separovic, F. and van Deventer, J. S. J. (2005a)."NMR study of structural ordering in aluminosilicate geopolymer gels." Langmuir 21(7):3028-3036.
16. Aiello R., Crea F., Nastro A., Subotiæ B., Testa F., (1991) Mechanochemical and Thermal Transformations of Amorphous and Crystalline Aluminosilicates, Zeolites 11(8) 767
17. Duxson P., Provis J.L., Lukey G.C., Mallicoat S., Kriven W.M, van Deventer J.S.J, (2005) Understanding the relationship between geopolymer composition, microstructure and mechanical properties Colloid Surf A - Physicochem Eng Aspect 269(1–3) 47.
18. Palomo, A; Blanco-Varela, M T; Granizo, ML; Puertas, F; Vazquez, T; Grutzeck, MW; (1999) Chemical stability of cementitious materials based on metakaolin, Cement Concrete Res. 29(7), 997-1004.
19. Xu, H. and van Deventer, J. S. J. (2000). "The geopolymerisation of alumino-silicate minerals." International Journal of Mineral Processing, 59(3): 247-266.
20. Xu, H. & Van Deventer, J.S.J., 2002, The effect of alkali metals on the formation of geopolymeric gels from alkali-feldspars, Colloids and Surfaces A - Physicochemical and Engineering Aspects, 216(1-3): 27-44.
21. Xu, H., van Deventer, J. S. J. and Lukey, G. C. (2001). "Effect of alkali metals on the preferential geopolymerization of stilbite/kaolinite mixtures." Industrial & Engineering Chemistry Research, 40(17): 3749-3756.
22. Lee, W.K.W. and van Deventer, J.S.J, 2002 "Structural reorganization of class F fly ash in alkaline silicate solutions." Colloids and Surfaces A - Physicochemical and Engineering Aspects, 211(1): 49-66.

23. Lee, W.K.W. and van Deventer, J.S.J., 2002 "The effect of ionic contaminants on the early-age properties of alkali-activated fly ash-based cements." Cement and Concrete Research, 32(4): 577-584.

24. Matthew Rowlesa and Brian O'Connor, (2003) Chemical optimisation of the compressive strength of aluminosilicate geopolymers synthesised by sodium silicate activation of metakaolinite J. Mater. Chem, 13 (5) 1161.

25. Duxsona P., Mallicoat S.W., Lukey G.C., Kriven W.M., van Deventer J.S.J., (2007) The effect of alkali and Si/Al ratio on the development of mechanical properties of metakaolin-based geopolymers, Colloids and Surfaces A: Physicochem. Eng. Aspects 29, 28–20.

26. Barbosa V.F.F., Mackenzie K.J.D., Thaumaturgo C., (1999) Synthesis and characterization of sodium polysialate inorganic polymer based on alumina andsilica, in: Geopolymer'99 Second International Conference, Saint-Quentin, France, 65–78.

27. Cioffi R., Maffucci L., Santoro L., (2003) Optimization of geopolymer synthesis by calcination and polycondensation of a kaolinitic residue, Res. Cons. Rec. 40, 27–38.

28. Hardjito D. and Rangan B.V., Development and properties of low calcium fly ash-based geopolymer concrete, (2005) Research Report GC, Faculty of Engineering, Curtin University of Technology, Perth, Australia, 1-130.

29. Gennaro R., Cappelletti P., Cerri G., Gennaro M., Dondi M., Langella A., (2004) "Zeolitic tuffs as raw materials for lightweight aggregates", Appl. Clay Sc. 25, 71.

30. ASTM Standards, (1983) ASTM Designation: C 109-80.

31. Perera D. S., Uchida O., Vance E. R., Finniem K. S., (2007) Influence of curing schedule on the integrity of geopolymers, J Mater Sci 42, 3099–3106.

32. D. Panias, I. P. Giannopoulou, T. Perraki, (2007) Effect of synthesis parameters on the mechanical properties of fly ash-based geopolymers, Colloids and Surfaces A: Physicochem. Eng. Aspects 301, 246–254.

33. Luz Granizo.M, Blanco-Varela M.T, Martı´nez-Ramı´rez S., (2007) Alkali activation of metakaolins: parameters affecting mechanical, structural and microstructural properties, J Mater Sci 42, 2934–2943

34. Lecomte.I, Henrist. C., Li´egeois M., Maseri. F, Rulmont. A, Cloots. R., (2006) (Micro)-structural comparison between geopolymers, alkali-activated slag cement and Portland cement, Journal of the European Ceramic Society 26, 3789–3797.

35. Andini S, Cioffi R, Colangelo F, Grieco F, Montagnaro F, Santoro L, (2008) Coal fly ash as raw material for the manufacture of geopolymer-based products, Waste Management 28, 416–423.

36. Mozgawa W, Deja J, (2009) Spectroscopic studies of alkaline activated slag geopolymers, Journal of Molecular Structure 924–926, 434–441.

37. Fernández-Jiménez A, Palomo A, (2003) Characterization of fly ashes. Potential reactivity as alkaline cements, Fuel 82, 2259–2265.

38. Criado M, Fernández-Jiménez A, A.G. de la Torre, Aranda M.A.G, Palomo A, (2007) An XRD study of the effect of the SiO2/Na2O ratio on the alkali activation of fly ash, Cement and Concrete Research 37, 671–679.

39. Davidovits. J, (2008) Geopolymer chemistry and applications. Morrisville, USA, p 570.

40. Kamseu E., Rizzuti A., Leonelli, Perera D., (2010) Enhanced thermal stability in K2O-metakaolin-based geopolymer concretes by $Al_2O_3$ and $SiO_2$ fillers addition, J Mater Sci 45, 1715–1724.

41. Xu H., Li Q., Shen L., Wang W., Zhai J., (2010) Synthesis of thermo stable geopolymer from circulating fluidized bed combustion (CFBC) bottom ashes, Journal of Hazardous Materials 175, 198–204.

42. Chindaprasirt P., Rattanasak U., (2010) Utilization of blended fluidized bed combustion (FBC) ash and pulverized coal combustion (PCC) fly ash in geopolymer, Waste Management 30, 667–672.

43. Xiao Yao, Zuhua Zhang, Huajun Zhu, Yue Chen (2009) Geopolymerization process of alkali–metakaolinite characterized by isothermal calorimetry, Thermochimica Acta, Volume 493, Issues 1–2, Pages 49–54.

44. De Silva P, (2008) Medium-term phase stability of Na2O–Al2O3–SiO2–H2O geopolymer systems, Cement and Concrete Research 38, 870–876.

45. Van Deventer J.S.J, Provis J.L, Duxson. P, Luckey G.C, (2007) Reaction mechanisms in the geopolymeric conversion of inorganic waste to useful products, J. Hazard. Mater. A139, 506–513.

46. Jadambaa Temuujin, Amgalan Minjigmaa, William Rickard, Melissa Lee, Iestyn Williams, Arie van Riessen, (2009) Preparation of metakaolin based geopolymer coatings on metal substrates as thermal barriers, Applied Clay Science 46, 265–270.

47. Xiaolu Guo, Huisheng Shi, Warren A. Dick, (2010) Compressive strength and microstructural characteristics of class C fly ash geopolymer, Cement & Concrete Composites 32, 142–147.

48. Yip C.K, Lukey G.C, Van Deventer J.S.J, (2005) The coexistence of geopolymeric gel and calcium silicate hydrate at the early stage of alkaline activation, Cement Concrete Res. 35 (9), 1688–1697.

49. Yip C.K, Van Deventer J.S.J, (2003) Microanalysis of calcium silicate hydrate gel formed within a geopolymeric binder, J. Mater. Sci. 38 (18), 3851–3860.