

Construction and Demolition Wastes as Resources for the Development of Green Building Materials

D. Kioupis, A. Skaropoulou, S. Tsivilis, G. Kakali

School of Chemical Engineering, National Technical University of Athens, Athens, 15773, Greece

Presenting author email: kioupid@gmail.com, tel.: +302107723284, fax: +302107723188

Abstract

This study deals with the alternative management of the construction and demolition wastes. In particular, it suggests the reuse of waste bricks and tiles as raw materials for the production of geopolymers which constitute a class of building materials with low environmental footprint. In order to evaluate the geopolymerization potential of the raw materials, alkali dissolution tests which determine the Si and Al leachability were conducted. The brick waste presented a slightly higher affection compared to the tile waste by the alkaline attack. In any case, the degree of dissolution is higher in NaOH than in KOH solutions. The geopolymer synthesis was optimized through the Taguchi multifactorial designing model of experiments based on the compressive strength of the final products. The results showed that the factors with the highest contribution on the mechanical behavior of the specimens are the alkalinity (R/Al) and the soluble silicon content ([Si]/R₂O) of the activation solutions. The delivery of the products with enhanced compressive strength values (41 MPa) is achieved for the following synthetic factor combination: [Si]/R₂O =1, R/Al=1.0, Na/Na+K=0.5, T=90°C and t =72h.

Keywords: CDWs, geopolymers, alkali activation, Taguchi.

1. Introduction

The increasing construction, renovation and demolition activities of the modern societies are accountable for the generation of one of the heaviest and most voluminous solid wastes, worldwide, called Construction and Demolition Wastes (CDWs). CDWs comprise a wide group of materials including concrete, bricks, tiles, gypsum, wood, glass, metals, plastic, solvents, asbestos and excavated soil.

Due to the rapid urbanization and industrialization, the CDWs landfill disposal poses additional environmental concerns. An effective way to release the environmental pressure is by recycling or reusing these materials. These actions reduce the landfill needed for CDWs disposal while avoid the consumption of non-renewable resources for new construction applications.

In the European Union (EU), the construction sector generates 531 million tons of CDWs per annum representing, nearly, the one quarter of the existing waste materials in the world (Table 1) [1]. According to the Waste Framework Directive 2008/98/EC, the EU members are obliged to take all the necessary measures in order to reuse, recycle or recover a minimum of 70% (by weight) of the non-hazardous CDWs by the end of 2020 [2]. Despite its potential, the level of recycling and material recovery of CDW varies greatly (between 0% and over 90%) across the Union [1]. It is well understood that the alternative management of these solid wastes constitutes a modern and imperative need.

Table 1. EU construction and demolition waste quantity and recycling rates (European Commission, 2011)

Country	CDW*	Recycling %	Country	CDW*	Recycling %
Denmark	5.27	94	Malta	0.8	0
Estonia	1.51	92	Holland	23.9	98
Finland	5.21	26	Poland	38.19	28
France	85.65	45	Portuguese	11.42	5
Germany	72.40	86	Romania	21.71	0
Greece	11.04	5	Slovakia	5.38	0
Hungary	10.12	16	Slovenia	2.00	53
Ireland	2.54	80	Spain	31.34	14
Italy	46.31	0	Sweden	10.23	0
Latvia	2.32	46	England	99.10	75

Lithuania	3.45	60	EU-27	531.38	46
Luxemburg	0.67	46			

*Million tons

A considerable part of the CDWs such as bricks, tiles, concrete and ceramic waste are aluminosilicate materials and can be utilized as resources for the development of new green building materials called geopolymers or alkali activated materials [3-5]. Geopolymers are a class of inorganic materials which comprises a three-dimensional polymeric-type structure. They are formed by the reaction of an alkaline solution with an aluminosilicate material at ambient or slightly elevated temperature. The aluminosilicate source is usually an industrial mineral, waste or by-product where part of silicon and aluminum species are preferably located in amorphous phases [6,7].

Geopolymers have a very low embodied energy and CO₂ footprint, compared to conventional building materials and they also exhibit rapid mechanical strength development and durability in corrosive environments. However, their greatest advantage is that, based on the choice of the raw materials and the design of the processing, geopolymers can meet a variety of requirements. This flexibility of geopolymer synthesis is of great importance when products with specific properties are required [8-10].

Up till now, little has been done upon the exploitation of CDWs in the geopolymer production. Most studies investigate the preparation of geopolymeric structures by using different mix proportions of CDWs (brick, tile or concrete) [11,12] and other aluminosilicate materials (such as fly ash or metakaolin) [5,13,14]. Furthermore, many research groups focused on the optimization of CDWs geopolymer synthesis by investigating the independent effect of various synthetic factors such as the type and the concentration of the alkali activator, the curing conditions, the particle size of the raw materials [4,11,15] while others dealt with the assessment of high temperature behavior of CDW geopolymers [12, 16].

This paper presents a study upon the synthesis of CDW geopolymers by the application of pure waste bricks or tiles as raw materials. The geopolymerization potential of the raw materials was determined by Si and Al leaching tests. In order to develop CDW geopolymers, the synthesis was optimized by examining the combining effect of the alkali and soluble silicon content of the activation solution, the curing conditions (temperature and time) as well as the kind of the alkali metal (Na or K) on the mechanical properties of the produced specimens. The synthesis optimization was performed via the Taguchi experimental design method, which provides a simple and efficient tool for optimizing parameters in complicated systems [17]. Finally, the products were characterized by means of XRD and FTIR.

2. Experimental

In this study, waste bricks and tiles, collected from various demolished buildings of Attica (Greece), were used as the raw materials. The CDW bricks and tiles were pulverized in order to obtain a certain fineness ($d_{50} = 20\mu\text{m}$) prior to their application. Table 2 shows the chemical composition of the raw materials in the form of oxides and trace elements, as determined by using X-ray fluorescence spectrometry (XRF).

Table 2. The chemical composition of the raw materials (% w/w).

CDW	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	K ₂ O	Na ₂ O	SO ₃	TiO ₂	P ₂ O ₅	Cl	L.O.I.
Bricks	51.31	14.62	8.45	6.33	8.10	2.28	0.57	0.44	0.66	0.08	0.04	397.0
Tiles	57.47	15.93	5.96	7.43	3.22	3.39	0.79	0.17	0.67	0.13	0.03	47.00

The Al and Si leaching of the raw materials was conducted by mixing 0.5 (± 0.0001) g of solid with 20 ml of alkaline solution under continuous stirring. The concentration of the alkaline solution and the dissolution time were kept at 10 M and 24 h, respectively [18]. After filtering, the liquid part was diluted to 250 ml, the pH was adjusted to pH < 1 by adding concentrated HCl acid and Atomic Absorption Spectroscopy (AAS) was used in order to determine the Al and Si concentrations. The solid part was examined by means of XRD in order to evaluate the effect of Al and Si leaching on the structure of the starting material.

The alkaline solutions were prepared by dissolving NaOH (> 99%, CAS: 1310-73-2) or KOH (86%, CAS: 1013-58-3) anhydrous pellets in distilled water and adding soluble Si in the form of SiO₂ (50% in H₂O, colloidal dispersion, CAS: 7631-86-9). The activation solutions were stored for a minimum of 24h prior to use, to allow equilibrium. Then, the raw materials (waste brick or tile) and the activation solution were mechanically mixed (standard mortar mixer: Controls 65-L0005) to form a homogenous slurry which was transferred to 50x50x50 mm cubic molds and mildly vibrated. The

specimens were left at room temperature for 2h and then were cured at various temperatures (25, 50, 70 and 90°C) for 24, 48 and 72 h. Compression tests were carried out on a Toni-technik uniaxial testing press, 7 days after the specimens' preparation (load rate 1.5 kN/s, according to the EN196-1 requirements). For each synthesis, three specimens were prepared and tested under compression.

The first part of the experimental involved the conduction of preliminary experiments on the geopolymer synthesis by the method of changing "one factor at a time". The studied parameters were the molar ratio [Si]/Na₂O which determines the amount of soluble silicon in the activation solution, the molar ratio Na/Al related to the alkalinity of the initial solution and the mass ratio solids/liquids which affects the workability of the pastes. For the preliminary experiments, the specimens were cured at 70°C for 48h.

The geopolymer synthesis based on CDWs was optimized through the Taguchi multifactorial designing methodology. The synthetic parameters selected to be investigated were the soluble silicon in the starting mixtures expressed as [Si]/R₂O molar ratio (R: Na or K), the alkalinity defined by the alkali to aluminium molar ratio (R/Al) in the starting mixtures and the kind of the alkali metal (R: Na or K). The experimental design involved the variation of the above factors in three levels (L₉(3³) orthogonal array) selected according to preliminary experiments (Table 3). Furthermore, the effect of the curing temperature (T = 25-90 °C) and time (t = 24-72 h) was evaluated. The optimization process was based upon compressive strength results.

After conducting the experimental designing model, the collected data were statistically analyzed by means of ANOVA (ANalysis Of VAriance) in order to determine the percent contribution of each factor on the development of compressive strength of the produced geopolymers.

Table 3. Factors and their variation levels.

Factor	Level 1	Level 2	Level 3
[Si]/R ₂ O	0	1	2
R/Al	0.4	0.7	1.0
Na/(Na + K)	0.0	0.5	1.0

The phase distribution of the powders was investigated by X-ray diffraction (XRD), on a Bruker D8 ADVANCE x-ray diffractometer. The data were collected in a 2θ range 2-70°, with 0.01° step size and 1 sec per step and were evaluated using Diffrac.Eva v3.1 software. FTIR measurements were carried out using a Fourier Transform IR Spectrophotometer (Jasco 4200 Type A). The FTIR spectra were obtained by the KBr pellet technique, in the wavenumber range from 400 to 4000 cm⁻¹ and resolution 4 cm⁻¹. The pellets were prepared by pressing a mixture of the sample and dried KBr (sample: KBr equals to 1:200, approximately) at 7.5 t/cm².

3. Results and Discussion

3.1. Extent of the starting materials dissolution

Table 4 presents the concentration of Al and Si in the solutions (10M NaOH or KOH) in relation to the starting material (CDW brick and tile). These values are the measured concentrations, after the leaching of 0.5 g of the solid material and the dilution of the liquid to 250 ml. Figure 1 presents the percentage of dissolved Al and Si after leaching, for 24 h, in 10 M NaOH and 10 M KOH. Since the starting materials have a varying content of Al and Si, Figure 1 presents the percentage of the total Al and Si of the starting material that has been dissolved. In this way, it was possible to evaluate how extensively the starting material has been affected by the attack of the alkaline solution.

Table 4. Extent of Al and Si dissolution in relation to alkali metal selection (ppm)

Solution	CDW brick		CDW tile	
	Si (ppm)	Al (ppm)	Si (ppm)	Al (ppm)
10 M NaOH	37.1	14.66	27.1	11.02
10 M KOH	29.3	9.69	18.6	6.22

The results of the alkaline dissolution experiments showed that waste bricks and tiles are susceptible to alkaline attack and, therefore, possess good geopolymerization potential. The brick waste showed a slightly higher alkaliine dissolution. In any case, the degree of dissolution is higher in NaOH than in KOH solution which is in well accordance to the literature [19]. However, it must be noted, that the leaching behavior of Al and Si cannot predict by itself the quality of the produced geopolymers. Further experiments concerning the preparation and properties of geopolymers from these raw materials have to be conducted.

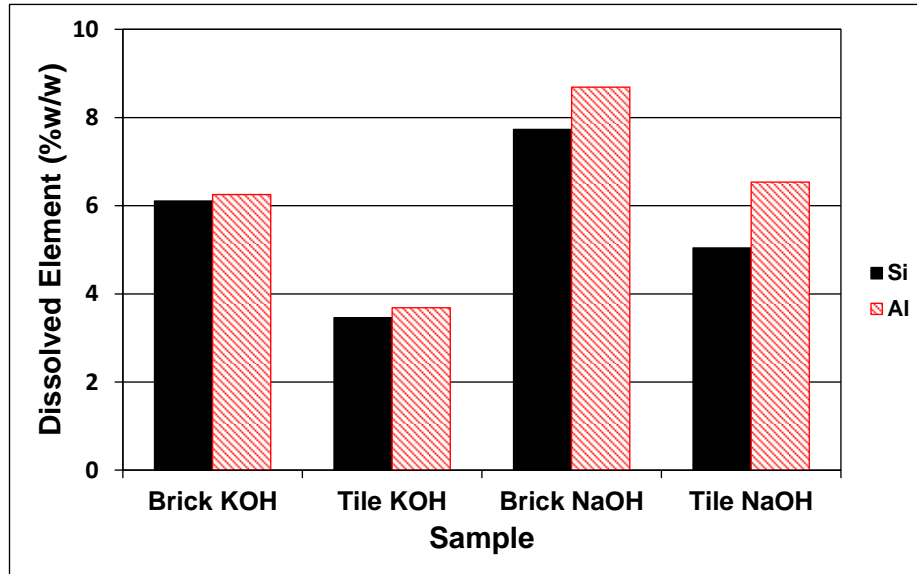


Fig 1. Dissolved Al and Si after leaching, for 24 h, in 10 M NaOH or KOH (% w/w)

Figure 2 presents the XRD patterns of the starting materials and their solid residues after leaching in 10 M NaOH for 24 h. The solid residues of both waste bricks and tiles contain the same crystalline phases as the starting materials but in lesser amounts as indicated by the decrease of the corresponding peaks.

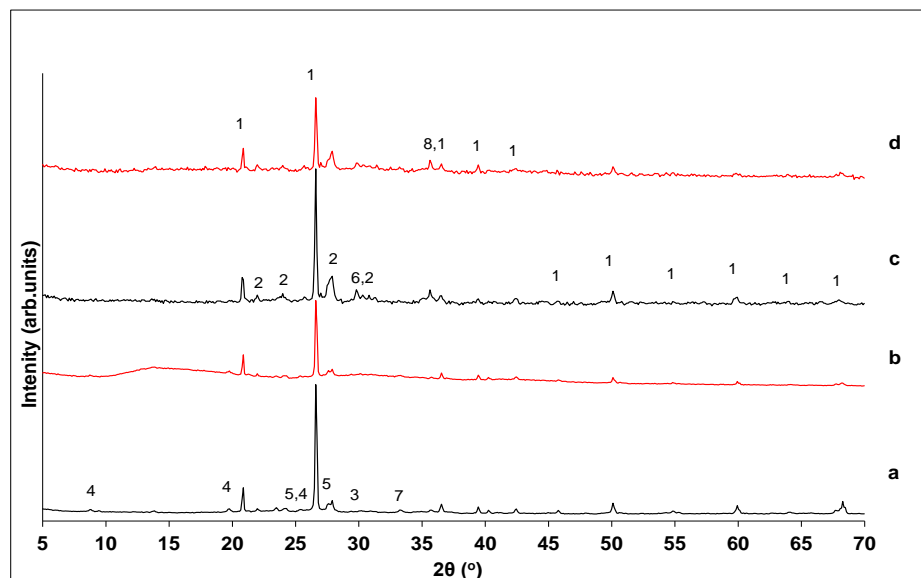


Fig. 2 XRD patterns of starting materials and solid residues.

a: waste brick, b: solid residue of waste brick after leaching in 10 M NaOH for 24 h, c: waste tile, d: solid residue of waste tile after leaching in 10 M NaOH for 24 h.

(1: quartz, 2: albite, 3: calcite, 4: muscovite, 5: microcline, 6: diopside, 7: potassium – calcium carbonate and 8: maghemite)

3.2. Preliminary experiments

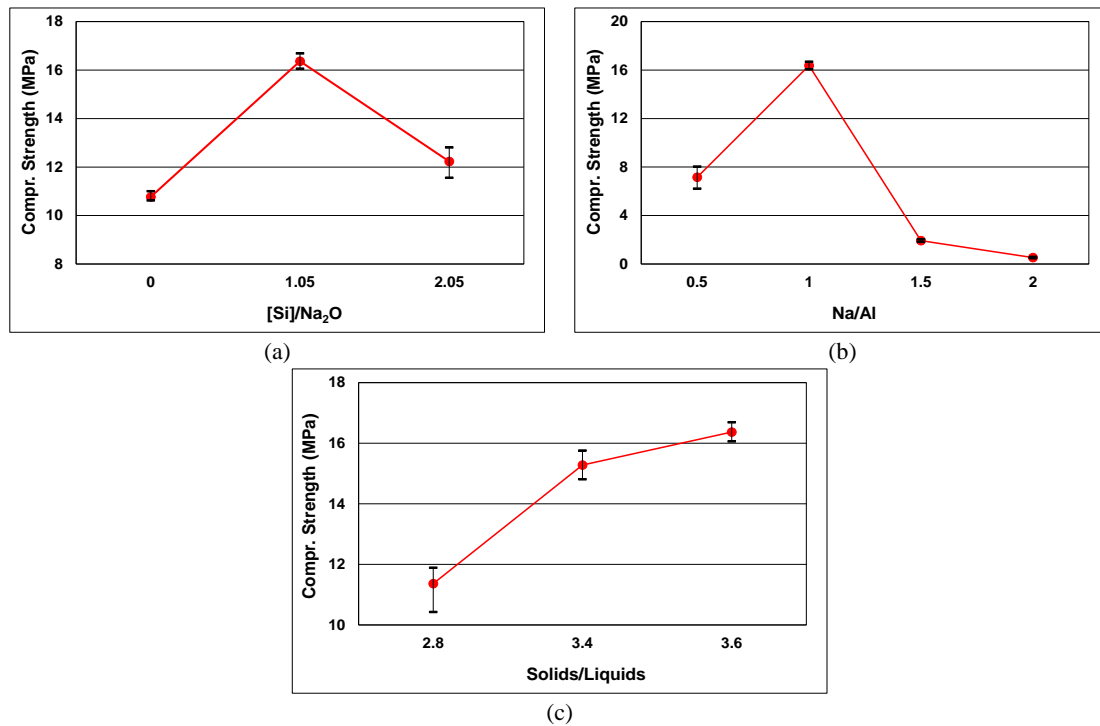
In order to evaluate the efficiency of the CDWs sources on the preparation of geopolymers, experiments concerning the synthesis and the properties of the geopolymers from CDWs were conducted and presented in the next paragraphs. Waste brick was qualified as the source material for the preparation of CDW geopolymers due to its higher Al and Si dissolution rates in relation to tiles as well as to the fact that is the most abundant aluminosilicate material in the CDWs [20].

Table 5 presents the preliminary experiments of the geopolymer synthesis based on waste brick conducted by changing “one factor at a time”. In Figure 3, the effect of the molar ratio [Si]/Na₂O (a), Na/Al (b) and mass ratio solids/liquids (c) on the compressive strength values is shown.

Table 5. Preliminary experiments by changing “one factor at time”

Experiment	[Si]/R ₂ O	R/Al	Solids/liquids
1	0.00	1.0	3.6
2	1.05	1.0	3.6
3	2.05	1.0	3.6
4	1.05	0.5	3.6
5	1.05	1.5	3.6
6	1.05	2.0	3.6
7	1.05	1.0	2.8
8	1.05	1.0	3.4

The results revealed that the presence of soluble silicon in the activation solution favors the production of geopolymers with enhanced mechanical properties. The soluble Si reacts with the Al of the raw material which is dissolved at the early stages of the dissolution process and forms oligomers which are polymerized to geopolymer structures [20]. The positive effect of the soluble silicon on the mechanical properties has an upper limit ($[Si]/Na_2O = 1.05$), higher than that a reduction of the compressive strength is observed. It seems that a high soluble silicon concentration in the activation solution slows down the dissolution of the Si of the aluminosilicate source and, therefore, delays the geopolymerization reactions.

**Fig. 3.** Effect of the synthesis factors on the compressive strength of the geopolymers.

The increase of alkalinity is initially beneficial for the development of compressive strength, since the alkali metals help the dissolution of the raw material and provide charge balance in the geopolymer network [21]. However, considerably high amounts of alkali metals do not further dilute the raw material, nor are incorporated to the matrix, on the contrary they favor the formation of carbonate phases which cause deterioration of the mechanical properties.

Finally, the effect of the water content (solids/liquids) on the geopolymerization of waste bricks was evaluated. As the solids/liquid ratio increases, compressive strength also increases, probably due to the increase of alkalinity in the activation solution. Solids/liquids ratios higher than 3.6 led to mixing problems and limited workability of the pastes. Therefore, it was impossible to deliver final products.

3.3. Synthesis optimization

In order to select the factors and the value ranges for the synthesis optimization through the Taguchi method, the observations of preliminary experiments conducted by changing “one factor at a time”

were taken into consideration. Table 6 presents the variation levels of the selected parameters of the Taguchi designing model along with the corresponding compressive strength values.

Table 6. Synthetic factors and compressive strength values for the produced geopolymers.

Sample	[Si]/R ₂ O	R/Al	Na/Na+K	Compressive strength (MPa)			
				(1)	(2)	(3)	Mean value
1	0.0	0.40	0.00	2.46	2.25	2.26	2.32
2	0.0	0.70	0.50	15.36	12.90	14.90	14.39
3	0.0	1.00	1.00	12.30	12.00	12.59	12.29
4	1.0	0.40	0.50	14.48	14.05	14.94	14.49
5	1.0	0.70	1.00	25.71	23.07	24.86	24.54
6	1.0	1.00	0.00	36.04	36.81	36.44	36.43
7	2.0	0.40	1.00	6.62	4.91	6.36	5.96
8	2.0	0.70	0.00	8.32	8.29	9.51	8.71
9	2.0	1.00	0.50	32.49	29.80	37.68	33.32

The effect of each parameter on the compressive strength is presented in Figure 4, which has been based on the processing of the data presented in Table 6. For example the compressive strength corresponding to Factor 1, Level 1 ([Si]/R₂O = 0.0) is the average strength of all samples containing [Si]/R₂O = 0.0. In the same figure, the impact of each factor on the compressive strength, estimated through ANOVA, is presented.

The factor with the major effect upon the development of the compressive strength of brick geopolymers is the R/Al molar ratio. An increase of this ratio from 0.4 to 1.0 results in the over-tripling of the compressive strength values (27 MPa).

The soluble silicon content ([Si]/R₂O) of the activation solution is the second more influential parameter.. An increase of the [Si]/R₂O from 0 to 1 favors the compressive strength, suggesting that the presence of soluble silicon is beneficial for the geopolymer formation. Further increase of soluble silicon ([Si]/R₂O > 1) led to the reduction of the strength of the specimens.

Concerning the alkali metal selection, the results showed that its contribution to the compressive strength is limited (7%). In any case, the use of a mixture of sodium and potassium (1:1) in the activation solutions marginally enhance the mechanical strength.

The results analysis showed that the optimal synthesis conditions for achieving the highest strength in the waste brick geopolymers are: [Si]/R₂O=1, R/Al=1.0 and Na/Na+K=0.5. The optimized value of the compressive strength for a 95% confidence interval was predicted to be 39.3574±4.7551MPa. In order to confirm the model prediction, geopolymer specimens with the optimized synthetic factors were prepared, appropriately cured for 7 days and subjected to compression measurements. The compressive strength (mean of 3 specimens) of the synthesis was found to be 40.98 MPa. This value falls within the predicting range, confirming in this way the validity of the used experimental design model.

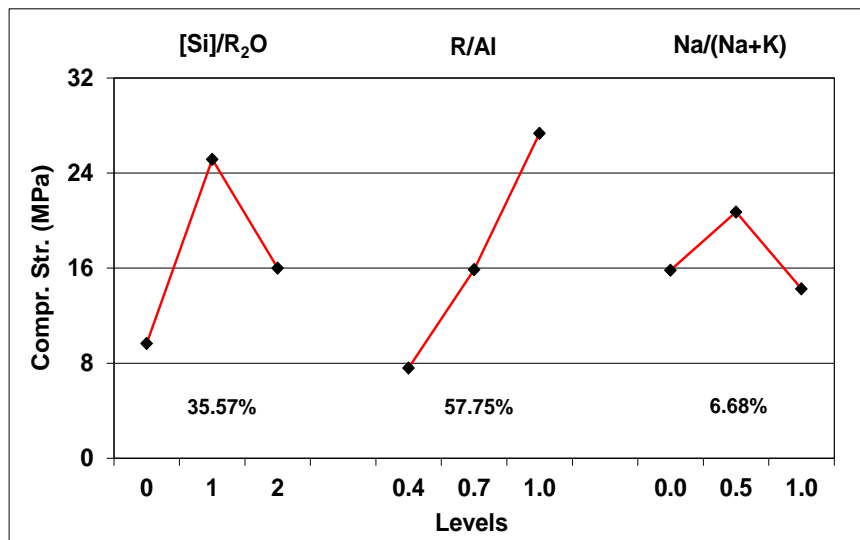


Fig 4. Effect of the studied factors on the development of the compressive strength of the specimens

3.4. Characterizations

The XRD patterns of the waste bricks and some representative geopolymers are presented in Figure 5. The main mineral phases in the CDW bricks are: quartz, albite, diopside, muscovite, microcline, maghemite and carbonates (calcium or potassium). In the case of geopolymers, the increase of the background, in the range 200-400 2θ , indicates the formation of the amorphous geopolymer matrix. When the soluble silicon content is low, the formation of zeolites, such as phillipsite, is favored and this is associated with lower compressive strength (Fig 5, exp. 4). On the other hand, the increase of soluble Si or the alkalinity in the activation solution, inhibits the formation of zeolites and favors higher compressive strength (Fig 5, exp. 9).

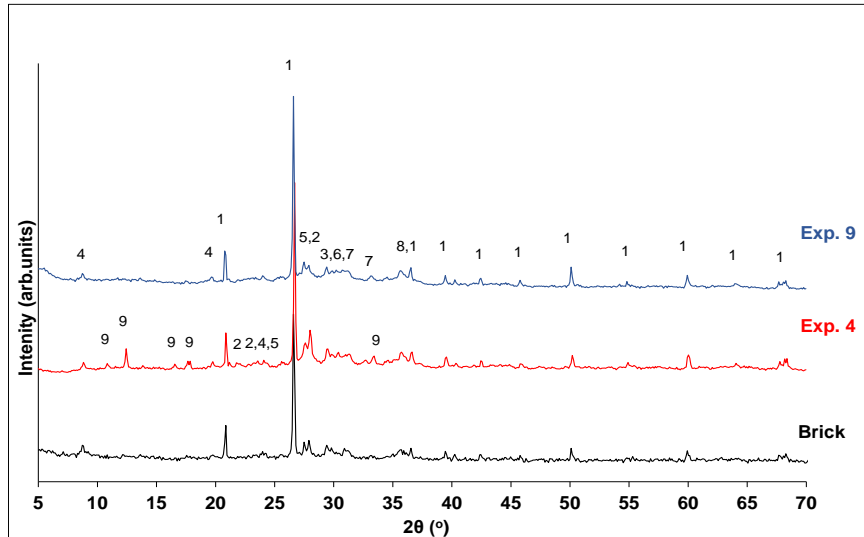


Fig. 5 XRD patterns of CDW brick and selected CDW brick geopolymers (1: quartz, 2: albite, 3: calcite, 4: muscovite, 5: microcline, 6: diopside, 7: potassium – calcium carbonate, 8: maghemite 9: phillipsite)

Figure 6 shows the FTIR spectra of the waste brick and a selected geopolymer sample. The region of interest in both the raw material and the brick geopolymers lies in the range between 800 and 1300 cm^{-1} . This broad hump exhibits overlapped peaks which are associated with the asymmetric stretching vibrations of Si-O-T (T: tetrahedral Si or Al) bonds. In the case of the CDW brick, the maximum of this hump is at around 1110 cm^{-1} while in the case of the geopolymers this hump becomes narrower and shifts to lower wavenumbers ($\sim 1000 \text{ cm}^{-1}$). This alteration is directly linked with the substitution of Si by tetrahedral Al and therefore the formation of an aluminosilicate network [22]. The Si-O-T asymmetric bending vibrations at $\sim 460 \text{ cm}^{-1}$ are not as sensitive as the stretching vibrations to changes of the aluminosilicate network.

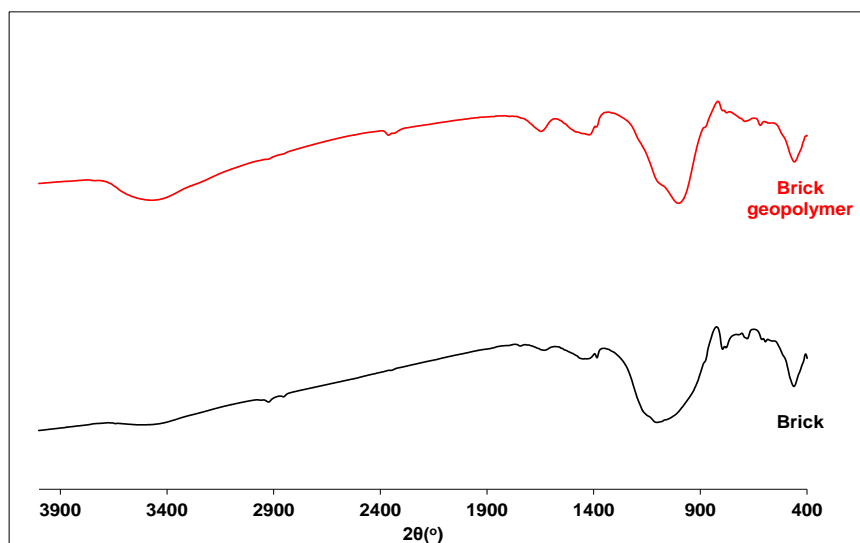


Fig 6. FTIR spectra of the raw material and brick geopolymer

3.5. Curing conditions

After the synthesis optimization of the waste brick geopolymers, the effect of curing conditions (temperature and time) on the compressive strength of the final products was investigated. For the preparation of the specimens the optimized factors as defined by the Taguchi analysis were applied. Figure 7 presents the compressive strength values of the geopolymers in relation to the curing temperature (25, 50, 70 and 90°C) and time (24, 48 and 72 h). An increase of curing temperature from 25 to 90°C leads to the drastic improvement of compressive strength by about 90%. In a similar manner, the curing of the specimens for 72 h achieves the maximum strengths. Based on these results, curing at 90 °C for 72 hours were selected as optimal conditions for the CDW brick geopolymer synthesis.

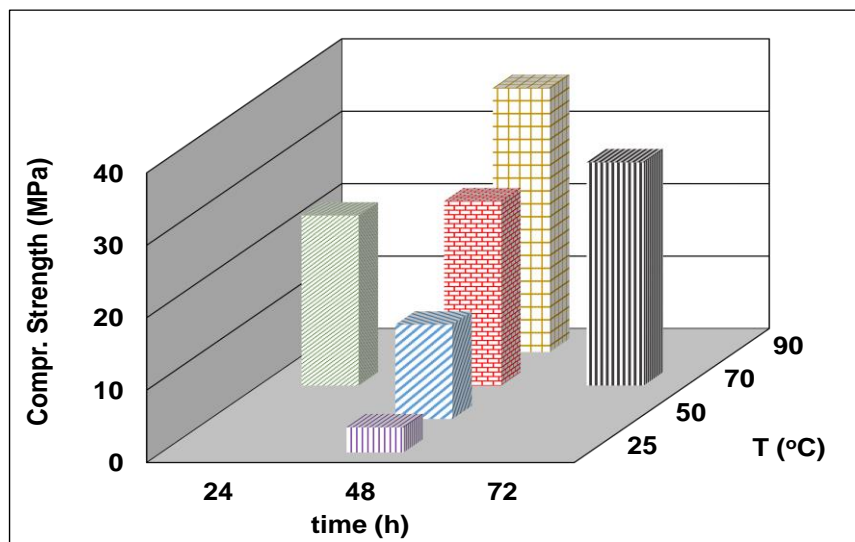


Fig. 7 Compressive strength values in relation to the curing temperature and time

4. Conclusions

This study suggests an alternative way for the valorization of the aluminosilicate CDWs (bricks and tiles) and their transformation to novel building materials through their alkaline activation. The final products (geopolymers) have significantly lower embodied energy and CO₂, compared with the conventional building materials.

Depending on the processing conditions, waste brick based geopolymers exhibit compressive strength from 2 to 41 MPa, indicating a wide application range for the alkali activation of CDWs.

The synthesis of waste brick based geopolymers can be optimized through the application of Taguchi multifactorial experimental design model. The synthesis conditions for the delivery of the highest strength products are: [Si]/R₂O=1, R/Al=1.0, Na/Na+K =0.5, T=90°C and t=72 h. The parameter having the highest impact on the development of the compressive strength is the alkalinity of the activation solution, with the quantity of the soluble silicon following in significance.

Acknowledgments

This research work has been financed by the European Union Horizon 2020 (H2020-EEB-2016-2017, “Green Integrated Structural Elements for Retrofitting and New Construction of Buildings – GreenINSTRUCT”, Grant No.: 723825).

References

1. European Commission (DG ENV), Final Report Task 2Service Contract on Management of Construction and Demolition Waste, 2011 (ENV.G.4/FRA/2008/0112).
2. Del Rio Merino, M., Navarro, J., Saez, P.: Legal Aspects which Implement Good Practice Measures in the Management of Construction and Demolition Waste. Open Constr. Build. Technol. J. 5, 124 – 130 (2011)

3. Komnitsas, K., Zaharaki, D., Vlachou, A., Bartzas, G., Galetakis, M.: Effect of synthesis parameters on the quality of construction and demolition wastes (CDW) geopolymers. *Adv. Powder Technol.* 26, 368–376 (2015)
4. Reig, L., Tashima, M.M., Borrachero, M.V., Monz, J., Cheeseman, C.R., Pay J.: Properties and microstructure of alkali-activated red clay brick waste. *Constr. Build. Mater.* 43, 98–106(2013)
5. Vásquez, A., Cárdenas, V., Robayo, R.A., Mejía de Gutiérrez, R.: Geopolymer based on concrete demolition waste. *Adv. Powder Technol.* 27, 1173–1179(2016)
6. Davidovits, J.: Geopolymers: inorganic polymeric new materials. *J. Therm. Anal.* 37, 1633–1656 (1991)
7. Provis, J.L., Palomo, A., Shi, C.: Advances in understanding alkali-activated materials. *Cem. Concr. Res.* 78, 110–125 (2015)
8. Duxson, P., Provis, J.L.: Designing precursors for geopolymer cements. *J. Am. Ceram. Soc.* 91, 3864–3869 (2008)
9. Provis, J.L., Bernal, S.A.: Geopolymers and Related Alkali-Activated Materials. *Annu. Rev. Mater. Res.* 44, 299–327 (2014)
10. Duxson, P., Fernández-Jiménez, A., Provis, J.L., Lukey, G.C., Palomo, A., Van Deventer, J.S.J.: Geopolymer technology: the current state of the art. *J. Mater. Sci.* 42, 2917 (2007)
11. Allahverdi, A., NajafiKani, E.: Construction Wastes as Raw Materials for Geopolymer Binders. *Inter. J. Civ. Eng.* 7, 154–160(2009)
12. Rapazote, J. G., Laginhas, C., Teixeira-Pinto, A.: Development of Building Materials through Alkaline Activation of Construction and Demolition Waste (CDW) – Resistance to High Temperatures. XII DBMC, Porto, Portugal(2011)
13. Hisham, M.K.: Effect of silica fume on the characterization of the geopolymer materials. *Inter. J. Adv. Struct. Eng.* 5, 1–10 (2013)
14. Ahmari, S., Ren, X., Toufigh, V., Zhang, L.: Production of geopolymeric binder from blended waste concrete powder and fly ash. *Constr. Build. Mater.* 35, 718–729(2012)
15. Pathak, A., Kumar, S., Vinay Kumar Jh.: Development of Building Material from Geopolymerization of Construction and Demolition Waste (CDW). *Trans. Ind. Ceram. Soc.* 73, 133–137 (2014)
16. Sun, Z., Cui, H., An, H., Tao, D., Xu, Y., Zhai, J., Li Q.: Synthesis and thermal behavior of geopolymer-type material from waste ceramic. *Constr. Build. Mater.* 49 281–287 (2013)
17. Panagiotopoulou, Ch., Tsivilis, S., Kakali, K.: Application of the Taguchi approach for the composition optimization of alkali activated fly ash binders. *Constr. Build. Mater.* 91, 17–22 (2015)
18. Panagiotopoulou, Ch., Kontori, E., Perraki, Th., Kakali, G.: Dissolution of aluminosilicate minerals and by-products in alkaline media. *J. Mater. Sci.* 42, 2967–2973 (2007)
19. Hu, X.U., Van Deventer, J.S.J.: The geopolymerisation of alumino-silicate minerals. *Int. J. Miner. Process.* 59, 247 (2000)
20. Oikonomou, N.D.: Recycled concrete aggregates. *Cem. Concr. Compos.* 27, 315–318 (2005)
21. Duxson, P., Mallicoat, S.W., Lukey, G.C., Kriven, W.M., van Deventer J.S.J.: The effect of alkali and Si/Al ratio on the development of mechanical properties of metakaolin-based geopolymers. *Coll. Surf. A: Physicochem Eng. Asp.* 292, 8–20 (2007)
22. Bakharev, T.: Geopolymeric materials prepared using Class F fly ash and elevated temperature curing. *Cem. Concr. Res.* 35, 1224–1232 (2005)