



Utilisation of Glass for the Production of Inorganic Polymeric Materials for Construction Industry

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How to cite this paper: Parisis, T., Mavrovounioti, N. and Antoniadis, P. (2020) Utilisation of Glass for the Production of Inorganic Polymeric Materials for Construction Industry. *Open Access Library Journal*, 7: e6123.

<https://doi.org/10.4236/oalib.1106123>

Received: January 30, 2020

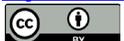
Accepted: April 20, 2020

Published: April 23, 2020

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Abstract

This paper deals with the utilisation of glass coming from municipal waste mainly for the production of inorganic polymeric materials, with advanced mechanical properties, intended for the construction industry. The development of glass-based geopolymers, achieving high compressive strength and low water absorption, is described, and the produced materials are compared with some common construction and building materials.

Subject Areas

Composite Material, Material Experiment

Keywords

Geopolymers, Construction Materials, Glass

1. Introduction

As it is well known, solid wastes cause severe contamination of the air, soil and surface and ground-water bodies. According to the EC Directive 96/61/EC [1], laid down for the management of wastes and by-products, their recycling, as well as their re-use as secondary raw materials, is strongly encouraged, in order to eliminate and/or prevent the environmental pollution, protect the human's health and preserve the natural resources. In this direction, an extended research, aiming at the development of viable technologies for the utilisation of industrial solid wastes and by-products in the production of useful materials, is performed during the last decades.

The geopolymerization technology [2] and therefore the study of the properties of the materials produced that are called geopolymers are an issue of

on-going extensive research worldwide. Geopolymers are a class of aluminosilicate-based materials with the potential to replace Portland cement in a variety of applications, particularly where chemical or thermal resistance is required [3]. Geopolymers are formed by the reaction between an alkaline solution and an aluminosilicate source. The hardened material has an amorphous to semi-crystalline three-dimensional structure similar to that of an aluminosilicate glass [4]. The reaction is exothermic and takes place at atmospheric pressure and temperatures below 100°C [2] [5]. The most proposed mechanisms for the geopolymerization process [5] [6] include the following four stages that proceed in parallel and thus, it is difficult to be distinguished: 1) Si and Al dissolution from the solid aluminosilicate materials in a strongly alkaline aqueous solution, 2) formation of oligomers (polynuclear hydroxy-complexes of silicon and aluminium) consisting of polymeric bonds of Si-O-Si and SiO-Al type, 3) polycondensation of the oligomers to form a three-dimensional aluminosilicate framework (geopolymeric framework) comprising of SiO₄ and/or AlO₄ tetrahedra linked alternately by sharing common oxygen ions and 4) bonding of the non-dissolved solid particles into the geopolymeric framework and hardening of the whole system into a final solid polymeric material.

Geopolymers possess excellent physicochemical and mechanical properties, like low density, micro- or nano-porosity, negligible shrinkage, high mechanical strength, thermal stability, fire resistance as well as chemical resistance, and notable surface hardness [2] [5] [7] [8] [9]. Due to these properties, geopolymers are often viewed as alternatives for certain industrial applications in the area of construction and building materials, since their properties are often similar to or even better than those of the conventional construction and fire protection materials. Apart from that, geopolymers seem to be more attractive than the conventional materials due to their low production cost, the lower energy requirements for production and the rational utilization of natural resources. This latter feature makes the geopolymerization technology very attractive as it achieves to turn numerous industrial by-products into added value materials [5] [7]-[13] and to offer possible technological solutions for the effective stabilization of hazardous wastes [14] [15], as well as for the encapsulation of toxic and radioactive wastes [6], contributing significantly to the elimination and/or reduction of the environmental pollution.

This paper aims at studying the geopolymerization of the glass coming from municipal wastes generated in order to develop novel inorganic polymeric materials for construction purposes. One of the main issues for the construction materials is the combination of high mechanical strength and low water absorption.

2. Experimental

2.1. Materials

The chemical analysis of the glass is given in **Table 1**. The X-ray Fluorescence (XRF) method (Spectro Xepos—Standardized for Aluminosilicate materials) was

used to determine the chemical composition of the glass. As shown in **Table 1**, the glass is very rich in silicon oxides and is relatively high in sodium, calcium and magnesium oxides.

The mineralogical analysis of the glass was performed according to the X-ray diffraction method on a SIEMENS D5000 diffractometer. As can be seen from **Figure 1**, the glass consists essentially of an amorphous silicate phase which is typical of glassy materials.

2.2. Synthesis of the Geopolymers

The following series of experiments were performed to study the behavior of glass geopolymers. It should be mentioned that NaOH was used as alkaline activator instead of KOH, and the reason is that the final application of the materials must show materials with high mechanical properties. NaOH as the most active solubilizes the amorphous Si and Al in the solid raw material to create a more stable mesh.

The first set of experiments concerns the study of the effect of alkali concentration on the strength of the finished material, with values of 6 M, 8 M, 10 M and 12 M. The purpose of the materials produced is to develop a compressive strength of at least 10 MPa, which is considered suitable for use as a sidewalk slab. The hardening temperatures of the geopolymers selected for experimental study in this series of experiments were 60°C. In all experiments, the relative maturation humidity of the geopolymers was 70%.

The methodology used to synthesize all the geopolymers used for the experimental study was as follows:

Initially, sodium hydroxide was dissolved in deionized water and the solid phase of the system (glass) was added in part and under constant mechanical stirring. The two phases were mechanically mixed for an additional 5 min, until a homogeneous slurry was created. Due to the low liquid phase content in the system, since the solid-to-liquid ratio (S/L) was $S/L = 4$ g/mL, the slurry generated was highly viscous. The synthesis followed is the one shown in **Table 2**.

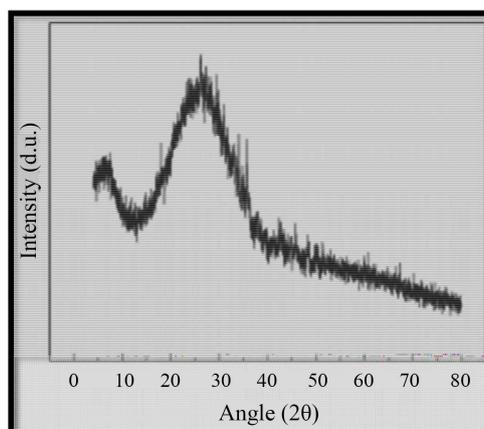


Figure 1. Mineralogical analysis of glass.

Table 1. Chemical analysis of glass.

Oxides	XRF Method
SiO ₂	68.9
Al ₂ O ₃	0.6
Na ₂ O	13.2
MgO	2.6
CaO	11.22

Table 2. Geopolymer synthesis conditions.

Synthetic Ingredient	Mass (g)	Content (wt%)
Glass	800.00	79.22
Sodium hydroxide	27.89	2.76
Deionized water	150.31	18.02
Total	1009.89	100

2.3. Formatting of Geopolymers

The glass was mixed with an alkaline sodium silicate solution (NaOH and SiO₂), used as an aqueous activator, in order to create a viscous paste that was subsequently molded in cubic 50 × 50 × 50 mm molds and cured in a laboratory-scale oven for 72 hours, under atmospheric pressure, temperature 60°C and relative humidity 70%. The formed cubic specimens were kept in closed molds for the first six hours of curing. Thereafter, they were demolded and left in the oven until curing. The cured specimens were left to cool for 48 hours at ambient conditions, before any test and analysis was carried out. The compressive strength according to ASTM C-109 and the cold water absorption according to the EN 771-1:2003 standard tests were determined.

2.4. Results and Discussion

After the geopolymer hardening, their apparent density, water absorption and alkalinity were tested. For the apparent density measurements, all three geopolymer specimens of each experiment were used. The water absorption test was subjected to one of the three geopolymer specimens of each experiment. The alkalinity of the geopolymers was measured in parallel with the water absorption, while the stability of the geopolymers was also studied in the same test.

Table 3 summarizes the results of the measurements of the properties of the geopolymers after their ripening at various temperatures.

As illustrated in **Table 3**, the apparent density of geopolymers is directly affected by alkalinity and increases as alkalinity increases. The water absorption of geopolymers by glass was extremely low compared to that of various conventional materials used today in the building materials and construction sectors. In the water absorption test, the geopolymers of all the experiments maintained their shape and consistency and did not undergo any external changes.

Table 3. Properties of geopolymers.

Alkalinity (M)	Density (Kg/m ³)	Water Absorption (%)	pH Initial Final	
6	1521.85	1.01	6.65	11.137
8	1591.00	0.84	5.55	11.557
10	1692.21	0.79	6.38	11.195
12	1734.42	0.77	6.24	11.104

The geopolymers of all the experiments showed high alkalinity and remained unchanged after being in water for 24 hours. In general, geopolymers are highly alkaline materials due to the high concentration of sodium hydroxide in their mass and the unreacted free alkali.

In addition of the synthesis conditions of the geopolymers followed in this experimental study, the appearance of the specimens is not directly affected by the alkali concentration.

2.5. Mechanical Strengths of Geopolymers

Upon the completion of hardening of the geopolymers (28 days) at the temperatures studied, two of the three geopolymer specimens of each experiment were subjected to uniaxial compression after being kept at ambient conditions for 24 hours. The average strength measurements of these specimens were considered as the ultimate mechanical strength of the geopolymers at the corresponding curing temperature.

As shown in **Figure 2**, the ultimate mechanical strength of the geopolymers is illustrated as a function of their alkalinity and their ultimate resistance in uniaxial compression appears to be substantially affected by alkalinity. Moreover, the increase in alkalinity results in a decrease—stabilization of the mechanical strength of the geopolymers and this was due to the excessive amount of NaOH in the system relative to the amorphous silicon and aluminium contained in the raw material. The alkalinity of 10 M is obviously the maximum amount of alkali that can dissolve the insoluble silicon particles in the glass.

More specifically, the total strength increased as the initial NaOH concentration increased from 6 M to 10 M, reaching a maximum value of 28 MPa. Further increase of the initial NaOH concentration to 12 M resulted in a decrease in the competitive force. Sodium hydroxide is concentrated in the alkaline wash of the glass in order to remove soluble amounts of silicon and aluminium from the glass. The rate of this divergence is directly related to the concentration of hydroxide ions in the liquid phase and generally increases with the increase of these ions. Increased concentrations of Si and Al in the liquid phase are necessary to maintain the oligomers of Si and Al complex species. The creation of such species accelerates the process of polycondensation which results in the development of complex polymer structures (**Figure 3**) that are important for the development of geopolymers. As shown in **Figure 4**, the increased SiO₂/Na₂O mass ratio in the liquid phase, promotes the polycondensation and polymer binder phenomena, thus, increasing the compressive strengths of the geopoly-

mers. An excessive amount of NaOH in the liquid phase reduces the $\text{SiO}_2/\text{Na}_2\text{O}$ mass ratio and inhibits the polymerization and shaping process of the polymeric binder by reducing the compressive strength of the geometric polymers.

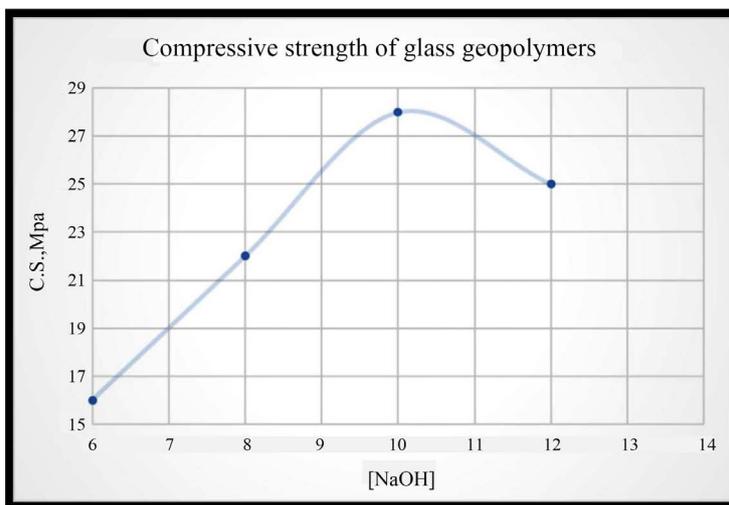


Figure 2. Resistance of geopolymers to uniaxial compression (MPa) as a function of alkalinity.



Figure 3. Glass based geopolymer.

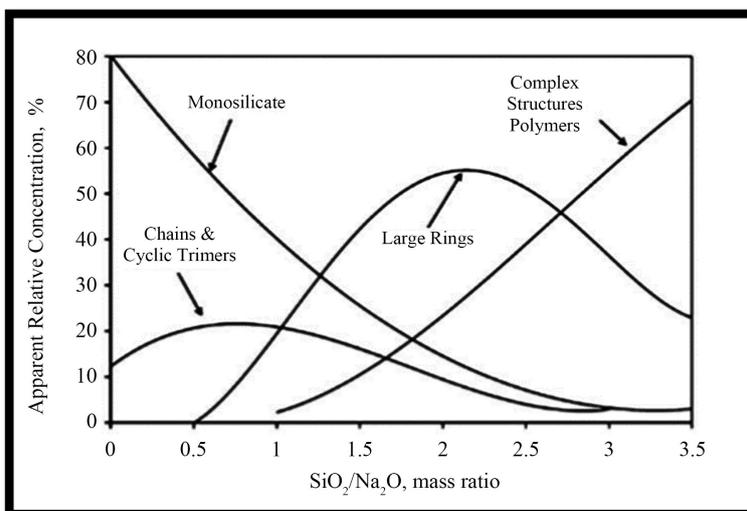


Figure 4. Structures depending on $\text{SiO}_2/\text{Na}_2\text{O}$ ratio.

In the uniaxial compressive strength test, the geopolymer specimens of all the experiments were not disassembled, confirming the existence of a stable, compact and cohesive structure, while behaving like natural rocks, forming a characteristic angle of 45° when they break.

3. Conclusions

In this work the development of added value geopolymer materials, using glass as raw material, is described.

1) Geopolymer material achieved a 28 MPa compressive strength with a <1% water absorption. This material has comparable or superior properties in relation to the commonly used construction materials (normal and high strength concretes).

2) The increase of alkalinity did not result in higher compressive strength.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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