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Effect of curing temperature on flexural properties of silica-based geopolymer-carbon reinforced composite

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Properties

ABSTRACT

Purpose: The aim of this paper is to find out the curing temperature at which we can achieve the best mechanical properties and adhesion between silica-based geopolymer matrix (Q1) and carbon HTS 5631 1600tex 24K fibre.
Design/methodology/approach: The carbon fibre was impregnated with silica-based geopolymer by means of home-made "impregnation machine". This equipment was designed based on simulating the real pultrusion or filament winding technique. Composite samples were made manually in silicon mould and cured under hot vacuum bagging technique at different temperatures. Flexural properties were determined under three-point bending mode in accordance with British Standard BS EN ISO 14125:1998. The sections perpendicular to fibres and surfaces of the composites were analysed by means of scanning electron microscope (SEM) to estimate the adhesion between geopolymer matrices and fibre reinforcement.
Findings: Relatively wide range of curing temperature from 70oC to 100oC at which we can obtain high flexural properties, maximal values of flexural strength 570 MPa, flexural modulus 65 GPa and relative deformation of composite was 0.98% when the composite was cured and dried at 75oC. Adhesion of the geopolymer matrix to carbon fibre was very good and hardly to determine the differences by SEM image observation within the range of optimal curing temperature.
Research limitations/implications: The curing time was too long to provide the geopolymerization process before it had been completed, this factor caused that it should be carried out in the future and we may use liquid absorption to determine how many cavities are in the composites.
Practical implications: The research presents original information on the influence of different curing temperatures on mechanical properties and micro-structure of silica-based geopolymer matrix – carbon composite. The results are useful for further investigations.
Originality/value: Determining the optimal curing temperature and micro-structure of silica-based geopolymer system to make it easy to find the curing time and other conditions to get the best properties of this type of materials.
Keywords: Silica-based geopolyme; Curing temperature; Lightweight; High strength; Fire resistant

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1. Introduction

The remarkable achievements in inorganic chemistry made through geopolymerization include mineral polymers which termed as polysiatate or geopolymers, making us possible to fabricate composite materials not only with excellent mechanical properties such as lightweight and high strength but also with ideal fire resistant (they can sustain temperatures up to 1200^{o}C with long term exposure), non toxic fumes and smokes, and resisting all organic solvents (only affected by strong hydrochloric acid) [1-6]. These special properties permit us to use more efficiently geopolymer matrix composites in high-tech technologies such as aerospace, naval architecture, ground transportation or automotive industry, especially for various applications that require high temperature resistance [1, 2, 5, 7,]. Conservative materials can be replaced efficiently by lightweight, high strength composites which are made from carbon or glass fibres and organic matrices or ceramic matrices (high costs associated with special processing requirements) and most organic matrix composites cannot be used in applications that require more than 200^{o}C temperature exposure) [1, 8, 9]. Moreover composites based on geopolymeric matrices are handled easily and do not require high heating, they are fabricated almost at room temperature or thermoset in a simple autoclave (usually under 150^{o}C) for several hours. In addition, most of types of fibres can be used with the geopolymer matrices and special ones can protect carbon from oxidation [5, 7]. In comparison with aluminium matrix – textile-reinforcement carbon fibre composite as an example, the composites were fabricated with the aid of a quite complex gas pressure infiltration technique, temperature of the process reached over 700^{o}C (because of the liquidus of aluminium temperature) and gas pressure was 90 Bar; long stay of carbon fibres in this temperature can cause degradation of the fibre, and hence, the properties of the composite [10]. Due to outstanding advantages of geopolymers more and more public and private research institutes and companies are investigating and finding applications in all fields of industry, such as civil engineering, plastics industries, waste management, automotive and aerospace industries, non ferrous foundries and metallurgy, etc [1, 5, 7, 8].

There are many factors that affect the geopolymerization process and mechanical properties finally, such as starting material including chemicals and mineral additives, alkali activators, plasticizers; processing conditions (usually time and temperature) [1, 6, 10-12]. The curing temperature is considered as an unconfined factor when researching compressive properties of geopolymer concretes [13, 14], geopolymer cement [15, 16] and fly ash-based geopolymer materials in general [17-19]. Finding out the curing temperature at which we can achieve the best mechanical properties and the adherence between silica-based geopolymer matrix (Q1) and unidirectional carbon HTS 5631 1600tex 24K fibre are the main targets of this paper.

2. Materials and Experimental parts

2.1. Matrix and reinforcement

The formulation of geopolymer matrix, which was abbreviated as "Q1", composed of thermal silica, kaolin and potassium water glass. Unidirectional carbon fibre HTS 5631 1600tex 24K was used as reinforcement with mechanical properties in Table 1 [20]

Table 1.
Properties of unidirectional carbon HTS 5631 1600tex 24K fibre

Kind of fibre	Average diameter [μm]	Linear density tex [g/km]	20^{o}C		
			A [%]	Rmo [MPa]	E [GPa]
Carbon HTS 5631 1600tex 24K	7	1600	1.84	3120	170

2.2. Fabrication of the geopolymer composites

The continuous fibres (roving) were impregnated with the geopolymer resin ("wet-out") by means of home-made "impregnation machine" (Fig. 1). This equipment was designed based on simulating the real pultrusion or filament winding technique. The velocity of the fibre during impregnation process was chosen based on the best penetration of geopolymer resin into the fibre; this value was around 34 m/h.

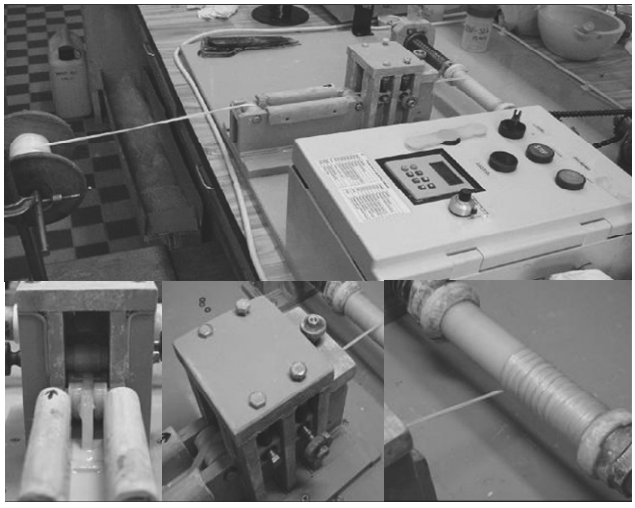

Fig. 1. Home-made impregnation machine

Impregnated fibres (pre-preg) were set into silicon moulds layer by layer.

The samples were cured under a technique called "vacuum bagging" (-1atm) at first in room temperature for 1 hour and then under a technique called "hot vacuum bagging" at different temperatures 55^{o}C, 65^{o}C, 75^{o}C, 85^{o}C, 95^{o}C, 105^{o}C, 115^{o}C for 5 hours in the oven.

Finally, the samples were dried in the oven at the same temperature of curing for another 5 hours. The completed composites containing approximately 37 wt. % or 40 vol. % of carbon HTS 5631 1600tex 24K fibre.

In preparing and curing process the samples were weighted to calculate the weight percentage of fibres in impregnated, uncured, cured and dried composites.

2.3. Mechanical testing set up

Series of five samples were prepared by manual lay-up technique using 16 bunches of pre-preg fibre for each specimen. The sample flexural properties were determined under three-point bending mode in accordance with British Standard BS EN ISO 14125:1998 [21] The flexural tests were conducted over a simply supported span of 64 mm with a centre-point load by Universal Testing Machine (Hounsfield Test Equipment Limited, England), Model Type: H50K-S (maximum load of the sensor: 50.000N); The deflection control with a mid-span deflection rate of 2 mm/min., at ambient condition temperature about 22±2°C and relative humidity 70%.

2.4. Adhesion of matrix and reinforcement

The sections perpendicular to fibres and surfaces of the composites were inspected in scanning electron microscope (SEM) to estimate the adhesion between geopolymer matrices and fibre reinforcements on the base of failure patterns in samples after flexural tests.

3. Results and discussion

The σ_{fM} was computed using equation:

$$\sigma_{fM} = \frac{3FL}{2bh^2} \tag{1}$$

Where:

σ_{fM} is the flexural strength, in megapascals (MPa)
F is the maximal load, in newtons (N);
L is the span, in millimetres (mm);
h is the thickness of the specimen, in millimetres (mm);
b is the width of the specimen, in millimetres (mm);

The flexural modulus E_f is calculated from equation:

$$E_f = \frac{L^3}{4bh^3}\left(\frac{\Delta F}{\Delta s}\right) \tag{2}$$

Where:

E_f is the flexural modulus of elasticity, expressed in megapascals (MPa);
Δs is the difference in the beam mid-point deflection (mm) between s" and s', which correspond to the given values of flexural strain ε_f" = 0, 0025 and ε_f' = 0, 0005, by following equation:

$$s'' = \frac{\varepsilon_f'' L^2}{6h} \text{ and } s' = \frac{\varepsilon_f' L^2}{6h} \tag{3}$$

ΔF is the difference in load F" and load F' at s" and s'respectively.

Calculating the relative deformation (ε) or the strain in the outer surface of the specimen at maximal beam mid-point deflection as following equation:

$$\varepsilon = \frac{6sh}{L^2} \tag{4}$$

The results of the effect of curing temperature on flexural properties of silica-based geopolymer composite Q1-carbon 5631 1600tex 24K is compiled in Table 2. It is easy to notice that the properties vary a lot at different temperatures of curing; when we increased the temperature of curing the flexural strength of the composite increased, however when the temperature was over 100°C, on the contrary, the ductility went down noticeably; cured at 55°C the relative deformation was 1.49% while this value was around 0.9% when the composites were cured over 85°C to 115°C.

Table 2.
Flexural properties of composites with geopolymer matrix Q1 and the carbon fibre at different temperature of curing

Temp. of curing (°C)	σ_{fM} (MPa)	E_f (GPa)	Relative deformation (%)	Density of composite (g/cm3)
55	386.42	45.01	1.49	1.82
65	425.25	49.10	1.43	1.85
75	570.72	64.31	0.98	1.86
85	504.71	62.56	0.88	1.79
95	511.92	69.13	0.90	1.78
105	361.49	65.21	0.82	1.65
115	98.64	20.19	0.78	1.32

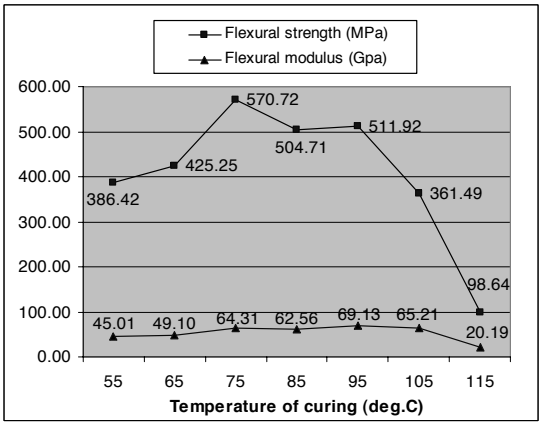

Fig. 2. Effects of temperature of curing on flexural strength and modulus of geopolymer composite Q1-carbon

We see the relatively wide range of curing temperature from 70°C to 100°C to get high flexural properties, at this temperature of curing, the flexural strength (σ_{fM}) of the composite fluctuated over 500 MPa, bending modulus (E_f) varied from 62.56 GPa to 69.13 GPa and relative deformation (ε) was about 1%; the maximal values of flexural strength was 570 MPa, flexural modulus was 65 GPa and relative deformation of composite was 0.98% when the composite was cured and dried at 75°C. The results can be comparatively compared with the best flexural properties of geopolymer matrix – unidirectional carbon fibre reinforced composite, σ_{fM} 527 MPa, E_f 84,95 GPa and bending relative deformation ε 0.65%, however, this composite was prepared using a standard vacuum bagging technique and heat press at 80°C and 3 MPa [22].

To study the adhesion between the carbon fibre and geopolymer matrix, the SEM images were investigated (Fig. 3). From the pictures at large magnification we saw that the adhesion was very good and it was difficult to recognize the differences between the pictures and say which temperature of curing is the best for the interaction between fibre and matrix.

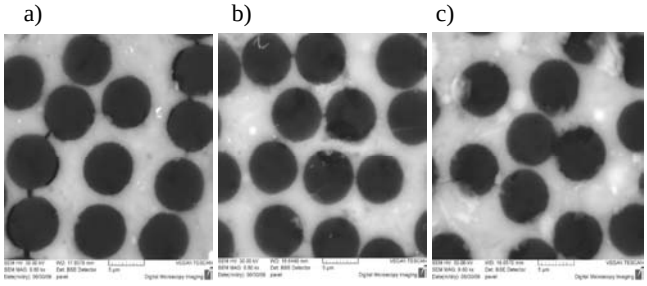

Fig. 3. SEM images of perpendicular sections of geopolymer composite Q1-carbon a) at 55°C, b) at 75°C and c) at 115°C curing temperature with magnification 9800x

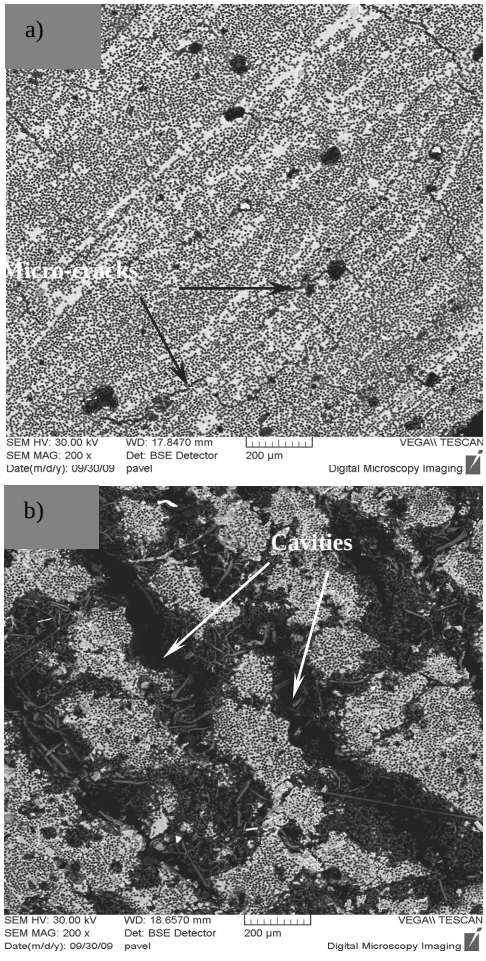

Fig. 4. SEM images of perpendicular sections of geopolymer composite Q1-carbon a) at 75°C and b) at 115°C curing temperature with magnification 200x

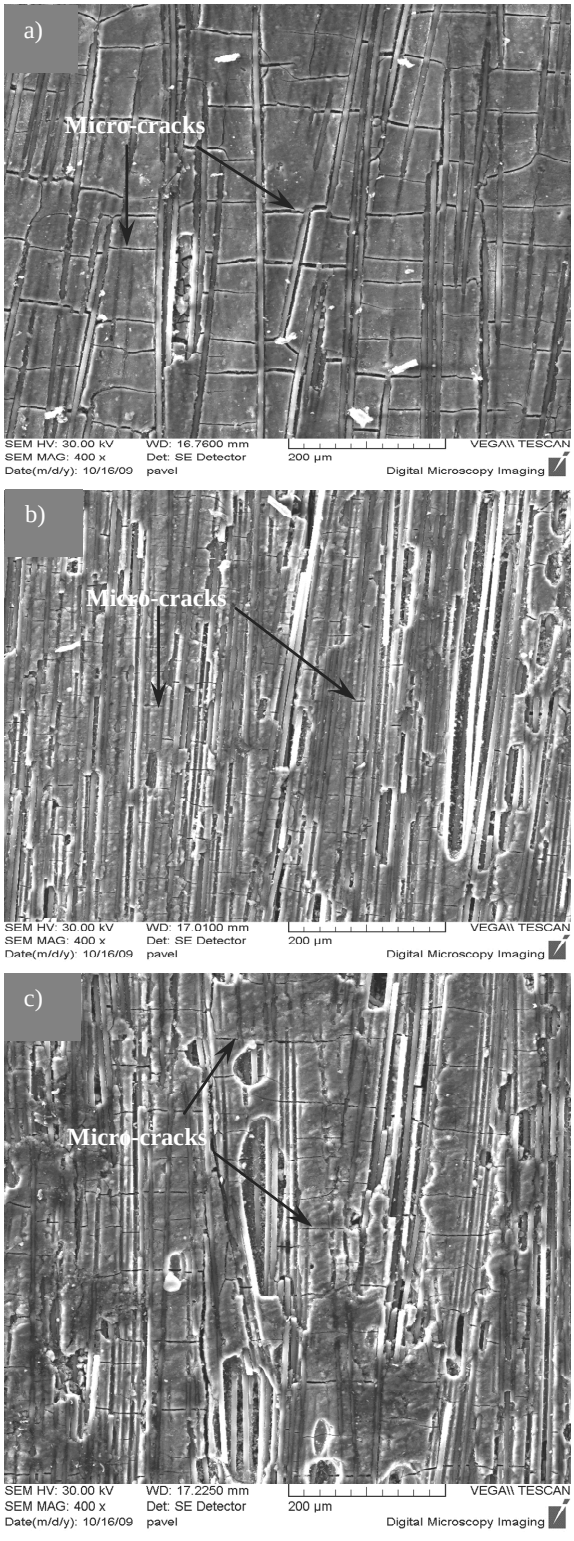

Fig. 5. SEM surface images of geopolymer composite Q1-carbon a) at 55°C, b) at 75°C and c) at 115°C curing temperature with magnification 400

However, with lower magnification 200x (Fig. 4), we can see that when the composite was cured at temperature higher than 100°C, the internal structure of the composites was deteriorated, many cavities were formed during geopolymerization; that made the density of composite decreased and the strength went down significantly (Table 2.). Cavities forming can be explained: water is released during the chemical reaction that occurs in the formation of geopolymers and when the temperature of curing was higher than 100°C the outer layers of composite were cured, curing more quickly and prevented water expelling from the geopolymer matrix when curing and then drying periods from escaping. This water boiled out at higher temperature and made the volume of composite increased forming cavities.

By observing SEM images of the composite surface (Fig. 5), we can see that when we increased the curing temperature, the micro-cracks seem smaller. The possible influence of micro-cracks for composites cured at 55 – 65°C on higher relative deformation of these samples. We can probably state that there is no firm bonding of matrix with fibre along the whole length of these samples. Therefore, higher curing temperature improved the geopolymerization process resulting in higher flexural strength. But over 100°C of curing temperature caused cavity generation, therefore a curing temperature of about 70°C to 100°C is recommended in practical applications for this silica-based geopolymer system.

All five beam samples failed by tearing of fibres. This is a very important finding, because the most common failure pattern reported in literature is the failure initiated by delamination or interlaminar shear fracture (Fig. 6) [1]. That also means the adhesion of the geopolymer matrix to the carbon fibre was very good.

Fig. 6. Typical failure pattern of the composite samples

Moreover, by means of SEM technique we can determine the micro-cracks as inborn defects in inorganic matrix composites. Further investigation should be concerned with this method if we would like to improve the mechanical properties of this composite by embedded ductile fillers.

4. Conclusions

The excellent properties such as lightweight, high strength, fire resistance, while evolving no toxic fumes and smokes, etc. permit us to think how to apply silica-based geopolymer matrix composites in high technologies such as aerospace, naval architecture, ground transportation or automotive industry, especially for applications that require high temperature sustainability.

The proper temperature of curing process for achieving good mechanical properties of this kind of silica-based geopolymer - carbon reinforced composites varies in relative large range, from 70°C to 100°C and at 75°C the composite achieves the maximal flexural strength about 570 MPa, bending modulus about 64 GPa and relative deformation 0.98%.

The SEM pictures and failure pattern indicate that the adhesion between geopolymer matrix and carbon fibre is very good; the images are the same, independent of the curing temperature inside the optimal range and we can say that the adherence of silica-based geopolymer matrix to carbon fibre and is better than metakaoline MK-750-based geopolymer with carbon fibre [1]. The micro-cracks in the matrix are determined as inborn defects of inorganic matrix composites.

Research and experiments are in progress in Department of Material Science, Faculty of Mechanical Engineering, Technical University of Liberec, and Research Institute of Inorganic Chemistry, Inc., Ústí nad Labem, The Czech Republic to improve properties of silica based geopolymer matrix by changing ratio of chemical composition, adding plasticizers and using it as matrix for composites with commercial fibres such as carbon, basalt and E-glass. The final purpose is to find out potential applications in industries.

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