The mechanical properties of flax and hemp fibres reinforced geopolymer composites

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Abstract. The article aim is to analyse the influence of addition of different kinds of flax and hemp fibres on the mechanical properties of the geopolymer composites. The paper describes the mechanical properties of fly-ash based geopolymer reinforced with short natural fibres made from the plants that have grown in Poland. The samples were prepared using sodium promoter and fibres (1% by mass of the composite). The empirical part of the research is based on the compressive strength tests, flexural strength tests and detailed microstructure examination. The research involved the samples reinforced by different kind of fibres - the flax and hemp fibres were prepared by different kind of treatment before addition to composites. The results show the influence of previous treatment of the fibres on the mechanical properties of the composites.

1. Introduction

The research on the composites based on geopolymers reinforced with the natural fibres is rather new area of investigation connected with the development of sustainability policy [1, 2]. Nowadays, there is the tendency to develop new and eco-friendly composites to replace the technologies based on non-renewable resources and technologies that are energy consuming such as Portland cement technology [3, 4]. The most promising alternative solutions for construction industry are technologies based on alkali-activated and/or geopolymer composites reinforced with fibres [5, 6]. The addition of artificial fibres usually makes mechanical properties better than the addition of natural ones [2], but from the environmental point of view, the addition of renewable fibres is more beneficial. The replacement of the artificial fibres with their natural counterparts decreases significantly CO2 emission calculated on the product – ecological footprint [7, 8].

The most important research on natural fibres as a reinforcement for geopolymers matrix was made with following fibres [1, 2]: cotton fibres [2, 9], coir [2, 10], sorghum fibres [11], wool fibres [12, 13], raffia [1], jute [14, 15], fique / sisal fibres [2, 16], corn husk [17], abaca [18], rice stem [19], pineapple leaf fibres [6] and bamboo [21]. The research results show that natural fibres can be a valuable additive for geopolymer composites and there is a high chance that they find the application as materials for construction, energy and other industries in the nearest future [15]. In comparison to materials reinforced by synthetic fibres they have numerous of advantages such as [2, 3]:

- Lower density than synthetic fibres.
- They are no toxic.
- They are easy for processing.
- The cost of production is lower than synthetic fibres.
They have high strength-to-mass ratio in early stage.
They have good tensile strength.
They are environmentally friendly, especially renewable and recyclable.

Some research has been made on flax and hemp fibres [4, 7]. The authors present that this kind of fibres can be used for the geopolymer composites, because they are relatively resistant to the alkaline environment of the matrix [7, 21]. The mechanical properties of such composites are better than unreinforced geopolymer matrix [7, 21]. Additionally, the reinforced composites show graceful failure, unlike the brittle failure of the matrix. In case of flax, the research shows that the composites are also thermally resistant up to 400°C (the flax fibres are protected by geopolymer matrix) [4, 7].

The flax and hemp fibres were selected to be investigated as they are easily available/reachable in Poland, due to the suitable climate and environmental conditions. The mechanical characteristics of mentioned fibres are suitable to be used in building materials [22, 23]. Moreover, the ecological footprint reduction is possible only when locally available raw materials are used. Thus, the transport of raw materials can be significantly reduced, with positive impact on environment. It has also justification on the economic site – the elimination of the cost of transportation [24, 25].

2. Experimental procedure
Specimens with 1% by mass of natural fibres were investigated. The variables include different types of fibres: flax and hemp.

2.1. Materials

2.1.1. Geopolymer matrix. The geopolymer matrix was prepared from fly ash from Skawina, Poland CHP plant and sand in ratio 1:1. This kind of fly ash is suitable for manufacturing geopolymer because of its physical properties and chemical composition. It has been confirmed by previous research, including SEM observations, EDS analysis, analysis of chemical composition (elements as well as oxides) and physical properties such as density [1, 2]. This fly ash form CHP plant in Skawina (Poland) is rich in oxides such as SiO₂ (47.81%), Al₂O₃ (22.80%), which is advantageous for production of geopolymers. The fly ash particles morphology was typical for such by-products of coal combustion and suitable for the process of alkali-activation [1, 2].

2.1.2. Flex fibres. As an additive, three kinds of flax fibres were applied: two kinds of flax tow and flax hurds. Tow is a coarse, broken fibre, removed during flax processing. They are usually shorter than 30 cm and are semi-products for textile fibres production. The hurds, called sometimes as hards, are the coarse parts of flax that adhere to the fibre after it is separated. They are usually residual from fibre production process. The following flax fibres were applied to compositions:

- Tow flax fibres after dew retting process (Figure 1) – the fibre is rather thin, but with some small hard part of stiffness elements (similar to flax hurds).
- Flax hurds (Figure 2) – the dimension is usually between 3-7 mm. The parts are stiff and ‘hard’.
- Green tow flax fibres (Figure 3) – the fibre is similar to tow flax fibre after dew retting process but is more stiff.

The fibres have been purchased from the Institute of Natural Fibres and Medicinal Plants in Poland.
2.1.3. Hemp fibres. As an additive, three kinds of hemp fibres were applied: they were in different forms and had different level of processing. The following hemp fibres were applied to compositions:

- Hemp hurds (Figure 4) – the dimension is usually between 5–10 mm (a little bit larger than flax hurds). The parts are more stiff than other investigated fibres.
- Decorticated hemp fibres (Figure 5) – thin fibre dedicated for decorticated purpose, without residues.
- Hemp microfibres (Figure 6) – very thin fibre with small number of thicker residues.

2.2. Specimens

Samples were prepared using sodium promoter, fly ash, sand and various flax and hemp fibres (1%, by mass of the composite). The process of activation has been made by 8M sodium hydroxide solution combined with the sodium silicate solution (liquid glass at a ratio of 1:2.5). To produce geopolymers, flakes of technical sodium hydroxide were used and an aqueous solution of sodium silicate (R-145) which molar module was 2.5 and density was about 1.45 g/cm³. The tap water was used instead of the distilled one. The alkaline solution was prepared by means of pouring the aqueous solution of sodium silicate over the solid sodium hydroxide. The solution was mixed and left until its temperature became stable and the concentrations equalized, which took about 2 hours. The fly ash, sand, alkaline solution and fibres were mixed about 20 minutes by using low speed mixing machine (to receive the homogeneous paste). Next, it was poured into two sets of plastic moulds. The first set consisted of the moulds dedicated to undergo compressive strength tests and the second set consisted of the moulds dedicated to undergo flexural strength tests. The samples were hand-formed and then subjected to vibratory removal of air bubbles. Tightly closed moulds were heated in the laboratory dryer for 24h at 75°C. Then, the samples were unmoulded. They were investigated after 28 days.
3. Analytical procedure

Scanning electron microscope (SEM) type JEOL JSM 820 with EDS has been used for microstructure research. The research has been made for fibres and the samples previously broken during compressive or flexural strength tests. The samples were covered with a thin layer of gold with JEOL JEE-4X vacuum sputter. The investigations were made at various magnifications (between 20–100x).

Compressive strength tests were carried out according to the methodology described in the standard EN 12390-3 (‘Testing hardened concrete. Compressive strength of test specimens’), because of the lack of separate standards for geopolymer materials. The tests involved at least 6 samples. Tests were performed using the universal testing machine - single-point load (Instron type 4465). Samples used to the compressive strength test had cubic shape and dimensions (approx.): 50 mm × 50 mm × 50 mm.

Flexural strength tests were carried out according to the methodology described in the standard EN 12390-5 (‘Testing hardened concrete. Flexural strength of test specimens’). The calculations were based on following equation:

\[ f_{cf} = \frac{FL}{d_1d_2^2} \]

where:
- \( f_{cf} \) – compressive strength, MPa
- \( F \) – maximal load, N
- \( l \) – space between supporting points, mm (for conducted tests: 140 mm)
- \( d_1, d_2 \) – sample dimensions, mm

Samples used to the flexural strength test had prismatic shape and dimensions (approx.): 200 mm × 50 mm × 50 mm. Tests were performed using the universal testing machine - single-point load (Instron type 4465).

4. Experimental results

4.1. Microstructure research

The SEM observations were made for fibres as well as for composition reinforced by fibres. The images were made at various magnifications - between 20–100x, depending on the particular fibre. The observation of microstructure of composites gave a preliminary information about the coherency of fibres (filler) with the geopolymer matrix (Figure 7). The fibre distribution was also evaluated.

In the Figure 8, there is presented the morphology of the flax tow after dew retting process. This kind of fibre is built with some smaller fibres that can be separated during the composition manufacturing. Flax hurds (Figure 9) have other morphology. They are rather compact and do not split themselves during the mixing process and composite production.
The green tow flax fibre (Figure 10) has very interesting structure. On the one hand it is quite compact, on the other the fibre structure is rough. The rough structure should allow the good coherence with composite matrix.

The decorticated hemp fibres (Figure 11) and the hemp microfibres (Figure 12) have thin fibres in their structure. It can be advantageous for composite, but it also requires carefully mixing process to ensure proper fibres distribution in the matrix. Thin fibres have tendency to create the agglomerates.

The fibre distribution in the matrix heavily influences the properties of the specific composite, as the fibres aggregation can decrease its mechanical properties. In case of investigated composites, the fibres distribution was regular. The microstructural observation also allows to notice that the structure is coherent (good adhesion the fibres to the matrix, Figure 7).

4.2. Compressive strength test
The results of the compressive strength test are shown in Table 1. The results of both fibre reinforced and plain (without fibres) specimens are presented.
Table 1. Compressive strength test after 28 days.

| Sample                                      | MPa    | standard deviation |
|---------------------------------------------|--------|--------------------|
| Geopolymer without fibre                    | 42.70  | 3.38               |
| Geopolymer with tow flax fibres after dew retting process (1%) | 31.92  | 1.80               |
| Geopolymer with flax hurds (1%)             | 30.72  | 3.36               |
| Geopolymer with green tow flax fibres (1%)  | 33.46  | 3.12               |
| Geopolymer with hemp hurds (1%)             | 35.90  | 2.00               |
| Geopolymer with decorticated hemp fibres (1%) | 42.65  | 4.17               |
| Geopolymer with hemp microfibres (1%)       | 35.20  | 7.03               |

Better results were attained through reinforcement by hemp fibres than flax ones. The best results have been achieved for decorticated hemp fibres. It is more than 42 MPa and it is comparable with geopolymer without reinforcement. Despite the decreasing of compressive strength, this result seems to be promising for future applications. The results for different flax fillers are very similar. The differences are statistically not important.

4.3. Flexural strength test

The results from the flexural strength tests are shown in Table 2. The results of both fibre reinforced and plain (without fibres) specimens are presented.

Table 2. Flexural strength test after 28 days.

| Sample                                      | MPa    | standard deviation |
|---------------------------------------------|--------|--------------------|
| Geopolymer without fibre                    | 5.22   | 0.25               |
| Geopolymer with tow flax fibres after dew retting process (1%) | 4.67   | 0.06               |
| Geopolymer with flax hurds (1%)             | 5.83   | 0.80               |
| Geopolymer with green tow flax fibres (1%)  | 5.12   | 0.37               |
| Geopolymer with hemp hurds (1%)             | 5.28   | 0.27               |
| Geopolymer with decorticated hemp fibres (1%) | 6.09   | 0.13               |
| Geopolymer with hemp microfibres (1%)       | 6.01   | 0.63               |

Similarly to the compressive strength tests, better results were attained through reinforcement by hemp fibres than flax ones. Moreover, the results with hemp fibres are slightly better than for geopolymer matrix without reinforcement. The best results give thin fibres such as decorticated hemp fibres and hemp microfibres than thick fillers. The flexural strength is important parameter for application in building industry, especially for more advanced construction. The achieved results show that new composites are comparable with contemporary used materials such as cementitious materials.

5. Discussion

There is lack of literature about research on influence of different kind of hemp and flax fibres on mechanical properties of the composites. Some research on the short flax fibres admixtures has been made based on geopolymers made from kaolinite clay with different amount of flax fibres: 4, 7 and 10 wt.% [7]. The research results pointed out improving mechanical properties of geopolymers by fibre additives, especially flexural strength. However, some of them seem to include incoherence like increasing of the flexural strength from 5.8 MPa to 70.2 MPa in connection with 10% flex fibre addition [7]. Those values of flexural strength were much higher compared to the results presented in our manuscript, which could be due to the different research methodology. 70 MPa was probably achieved
at total failure of the sample and not at the “first crack”. Such test probably shows a flexural strength of fibres not of the whole composite. The other research is focused on flax fabric reinforcement [4].

There is also lack of complex research on hemp fibres. These fibres have been investigated in composites based for complex matrix that included: fly-ash, waste glass, ordinary Portland cement (OPC) and recycled aggregates [25]. The geopolymer mortar was reinforced by short hemp fibres (ca. 20–30 mm) up to 8% by mass of the composite Investigation shows decrease of mechanical properties caused by fibre additives (about 50% in the compressive strength due to addition of 8% hemp fibres). The compressive strength for the matrix was 45 MPa, after 8% fibre addition it was only 12 MPa. The similar situation was also in the case of flexural strength - it was 3.45 MPa for geopolymer matrix and 2.13 MPa for composite. Despite the decreasing mechanical properties for the composites, the authors stress the advantageous for the new material, especially environmental benefits – reduce the carbon footprint, connected with using the natural fibres (the use of at least 8% hemp fibres leads to carbon negative emissions -19.7 kgCO$_2$eq/m$^3$) [25]. The decrease of mechanical properties was attributed to the high content of the fibres. Limitation to 1 or 2% by mass of the composite will be more advantageous for mechanical properties.

6. Further research
The results of the research and literature review show that the geopolymer composites can be low-cost and effective construction materials. Further tests should be performed in order to optimize the mechanical properties of the composites as well as to investigate other properties such as absorptivity and resistance for different environments. The possible application for this kind of composites is construction industry, so the planned research should involve frost resistance as well as heat resistance and also durability issues. In the long perspective, it is also recommended to design some prototype elements, exemplary bricks and make the LCA analysis for the chosen products.

7. Conclusions
Geopolymer composites reinforced with various flex and hemp fibres have been produced and characterized. The samples were prepared using sodium promoter and six various types of fibre additives (1% by mass of the composite). The matrix was geopolymer cement (fly ash and sand in ratio 1:1).

The observed differences between the fibres influence the mechanical properties of the whole composite. The composites with hemp fibres outperformed the flex reinforced ones in terms of mechanical properties - higher values of both compressive and flexural strength were observed when hemp was involved. The best results were achieved in case of fine fibres such as decorticated hemp fibres and microfibres. This study shows that it is possible to produce the composites of reasonable properties from the industrial wastes (fly ash) and renewable resources – natural fibres.

Even the fibre addition does not improve the final strength of the composite significantly, the overall mechanical parameters are positively influenced; the presence of the fibres changes the character of failure mechanism from brittle to ductile. Additionally, using the natural fibres instead of industrial ones has positive impact on environment. Thus, the designed composites have potential to be used as new eco-friendly material e.g. in construction industry.

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