LIGHTWEIGHT COMPOSITES BASED ON CELLULOSIC MATERIAL

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Abstract: In this paper, the overview of knowledge about the properties of lightweight composites based on hemp hurds with MgO-cement as an alternative binder is given. There are summarized the results of the development of density, compressive strength, thermal conductivity, thermal behaviour and water sorption of the composites in the dependence on different filler portion and its mean particle length. Decrease in density, compressive strength and thermal conductivity coefficient with an increasing share of hemp hurds (20-60 vol. %) in hardened composites was found. Physical and mechanical properties of bio-composites depend also on the particle size in length dimension of hemp hurds. Compressive strength and water sorption of composites after 28 days of hardening increase with decreasing mean particle length, while this effect on thermal conductivity of composites was not confirmed. Thermal loading of composites (50-200°C) led to their damage by increasing temperature. The most significant decrease in density and compressive strength of composites was obtained at temperature of 200°C.

Key words: Lightweight composites, hemp hurds, MgO-cement, compressive strength.

1. INTRODUCTION

Production of building materials causes significant air pollution by CO₂ emissions. It is responsible for the consumption of energy from fossil fuels resources and the depletion of large amounts of non-renewable raw materials (Pacheco-Torgal and Jalali, 2011). In view of these facts, the construction industry is non-sustainable. Principles of sustainable construction of the buildings bring new requirements to develop sustainable materials. Actual strategic role in achievement of a sustainable building industry consists in the selection and rational use of material and energy resources and technologies by controlled minimization of the total production of CO₂ emissions as well as in meeting the requirements for the quality, reliability and functionality of building materials and structures and for provision of optimal indoor climate. One of the possible ways of sustainable alternatives development in the construction industry is moving from the limited and finite material resources to easily renewable raw material resources. In this respect, there is a large group of materials of plant origin as rapidly renewable raw materials which can be used as suitable reinforced component to lighten concrete mixtures. Nowadays, new bio-composites containing natural fibres and/or particles, lignocellulosic wastes are the subject of research, specifically in building materials preparing as reinforcement of composites for their installation in structures due to their many advantageous properties. They are low density materials yielding relatively lightweight composites, which can exhibit strength parameters, as shown in previous studies for the use of various natural fibres such as flax, cellulose, hemp, jute, sisal, kenaf, bamboo in composites (Almeida et al., 2009; Bismarrck et al., 2002; Bruijn, 2008; Ouajai and Shanks, 2009; Boghossian and Wegner, 2008). Whereas the use of cellulosic fibers in the polymer or cement matrix is known and it leads to comparable mechanical and insulating properties of the resulting environmental friendly bio-composites with composites based on conventional man-made fibers like glass and carbon fibers (Li et al., 2006; El-Tabwe, 2009), at present the attention is given to research of their incorporation in mineral matrix, such as lime or other alternative binders (mix of hydraulic lime, hydrated lime and pozzolanic admixtures) and to composite properties testing (Allin, 2005; Bevan and Woolley, 2008; Kidalova et al, 2012). Often, cellulosic materials are produced as waste by-products of industrial or agricultural processes. They in form of fibres or non-fibrous material are an ideal candidate for an effective reinforcement in concrete/mortars. One of the most important targets in recent materials research is their utilization into composite (Ahankari et al, 2011; Ashori and Nourbakhsh, 2010; Pacheco-Torgal and Jalali, 2011). Among a wide variety of plant sources, a great importance is attached to technical hemp (Cannabis Sativa) for its use in bio-composite (Arnaud and Gourlay, 2012; Colinart et al., 2012; de Bruijn et al, 2009; Shahzad, 2012). Specifically, hemp is very...
heterogeneous substance, its microstructure is extremely complicated and therefore cannabis itself can be regarded as a composite system (Brett and Waldron, 1996; Dai, 2010). The basic unit consists of cellulose polymeric chains aligned and gathered in microfibrils. They are linked to each other by lignin, pectin and hemicellulose. The strength and stiffness of the fibres are provided mostly by hydrogen bonds between the different chemical components. Thermal stability, resistance to UV attack or biodegradation of hemp depends on the hemp composition and individual properties of each component. Hemicellulose is responsible for the biodegradation, moisture absorption and thermal degradation of the fibre (Sahed and Jog, 1999). Lignin and pectin are thermally stable but are responsible for the UV degradation of the fibre (Kalia et al., 2011).

The effective utilization of hemp fibre, particles and/or yarn as a renewable source of cellulosic fibres in structural composite components depends on their important characteristics, mainly material parameters, physical and mechanical properties in relation to composite reinforcement. A major restriction in the successful use of natural fibers in durable composites applications is their high moisture absorption and poor dimensional stability (swelling). It needs to be considered because this hydrophilic behaviour affects the properties of the fibres themselves as well as the properties of their composites (Bledzki and Gassan, 1999). The durability of natural fibres reinforced composites may be limited due to their high permeability and lack of resistance to micro-crack growth (Savastano et al., 2009). In terms of mechanical properties of composite, the properties of binder and fibre/matrix interface are significant too (Madsen et al, 2012). The advantages and limitations of the cement-bonded composites made with lignocellulosic wastes are described in the review (Karade, 2010). Not mentioned in this work, hemp shivs (or hurds) like the chopped residues of the woody stem of the plant offer large possibilities of their use in the building construction (Arnaud and Gourlay, 2012; Bevan and Woolley, 2008; Stikute et al., 2011). Composite based on hemp is often called hemp concrete (or hempcrete). It is known hemp concrete can not be used for structural purposes. Many studies have showed that the mixtures manufactured from the raw materials (binder, hemp shivs, water) in various proportions cover a wide range of applications (Elfordy et al., 2008; de Bruijn et al., 2009; Nguyen et al, 2009).

Our previous research was oriented on utilization of unbleached cellulose fibers from pulping and hemp hurds slices with different binding agents (hydrated lime, cement and zeolite) and their combination into composites (Kidalova et al, 2011a, Kidalova et al., 2012). Alternative binder MgO-cement has been used for preparation of composites based on hemp hurds slices and technically significant properties of hardened composites were studied (Kidalova et al., 2011b, Kidalova et al., 2011c; Stevulova et al, 2012a; Stevulova et al; 2012b). In this paper, the physical (density, thermal conductivity, thermal behavior and water absorption) and mechanical properties (compressive strength) of the composites based on various contents of hemp hurds and mean particle length after 28 and 90 days of hardening are compared.

2. MATERIAL AND METHODS

In our experiments, the technical hemp hurds coming from the Hungary company (Hungarohemp Rt, Nagylak; bulk density = 115 kg.m$^{-3}$) was used. The quantity of components contained in hemp hurds was determined by extraction. The content of polysaccharide component (holocellulose) is 71.5 %. The amounts of cellulose and hemicellulose like holocellulose components are 44.3 and 27.2 %, respectively. Other components present in hemp are lignin (22 %), ash (1.3 %) and compounds soluble in toluene and ethanol (6.2%). Average moisture content in hemp sample is 10.78 %.

Three dried samples of hemp hurds slices were used for composite preparation. Original hemp hurds (sample 1) was polydisperse with a wide anisometric particle size distribution (Table 1). 94.1 wt. % of sample 1 had length longer then 1 mm and the longest slices had size of 8 mm. Two fractions (sample 2 and 3) were obtained by sieving the original hemp hurds to study effect of mean particle length on composite properties (Table 1). The thickness and width of all hemp particles of three hemp hurds samples have changed little.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Granularity, [mm]</th>
<th>Mean particle length, [mm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>8 - 0.063</td>
<td>4.29</td>
</tr>
<tr>
<td>2</td>
<td>8 - 4</td>
<td>7.42</td>
</tr>
<tr>
<td>3</td>
<td>4 - 0.063</td>
<td>2.33</td>
</tr>
</tbody>
</table>

Milled MgO (mean particle diameter: 6.85 μm) obtained by low thermal decomposition of magnesite (CCM 85, SMZ a.s. Jelsava, Slovakia), silica sand (Sastin, Slovakia) with SiO$_2$ content of 95 - 98% and sodium hydrogen carbonate are components of binder (MgO-cement).

The compositions of the experimental mixtures with different portion of hemp sample 1 are presented in Table 2. Mixtures I.1 and II.2 were prepared according to recipes published in (Bydzovsky, 2009) and (Allin, 2005), respectively. The mixtures I.2-I.3
and II.1-II.3 are modifications of previously mentioned mixtures. The composition of three experimental mixtures based on different mean particle length of hemp hurds corresponded to the formulation mixture I.2 (40 vol. % of hemp shives, 29 vol. % of MgO-cement and 31 vol. % of water). Designation of these mixtures is consistent with labeling of hemp hurds samples in Table 1.

<table>
<thead>
<tr>
<th>Mixture</th>
<th>Composition of mixture [vol.%]</th>
<th>Hemp hurds</th>
<th>MgO-cement</th>
<th>Water</th>
</tr>
</thead>
<tbody>
<tr>
<td>I.1</td>
<td>20</td>
<td>45</td>
<td>35</td>
<td></td>
</tr>
<tr>
<td>I.2</td>
<td>40</td>
<td>29</td>
<td>31</td>
<td></td>
</tr>
<tr>
<td>I.3</td>
<td>60</td>
<td>19</td>
<td>21</td>
<td></td>
</tr>
<tr>
<td>II.1</td>
<td>20</td>
<td>58</td>
<td>22</td>
<td></td>
</tr>
<tr>
<td>II.2</td>
<td>40</td>
<td>22.5</td>
<td>37.5</td>
<td></td>
</tr>
<tr>
<td>II.3</td>
<td>60</td>
<td>15</td>
<td>25</td>
<td></td>
</tr>
</tbody>
</table>

The fresh mixtures were prepared in the labour mixer and the standard steel cube forms with dimensions 100mmx100mmx100mm were used to preparation of specimens. Next two days the composites were taken out of the forms and cured under laboratory conditions according to standard procedures during 28 and 90 days. At the end of 24 h, the samples were removed from the mold, and composite bodies hardened under the conditions required according to standard (STN EN 206-1, 2009). The density, thermal conductivity coefficient, compressive strength and water absorption were measured on hardened composites. Density was determined in accordance with standard (STN EN 12390-7, 2011). The thermal conductivity coefficient was determined by using the commercial device ISOMET 104.

Compressive strength of the cube specimens under controlled conditions was determined as the maximum load per average cross-sectional area by using the instrument ADR ELE 2000 (Ele International Limited, United Kingdom). Water absorption was specified in accordance with the standard (STN EN 12087/A1, 2007). First all the specimens were dried in an oven at 70°C and then cooled and weighed. Water absorption was carried out by immersing the hemp composites in de-ionised water bath (PE closed container) at laboratory temperature. After one hour immersion, the specimens were taken out from water and all surface water was removed with a clean dry cloth. The specimens were reweighed, changes in dimensions were measured too and afterwards the specimens were dried in an oven at 70°C up to constant weight for following measurements. Thermal loading of 28 days hardened composites (recipe I.2) was carried out in an electric furnace at temperatures of 50, 100 and 200°C in period time 48 hour or 72 hour. Influence of thermal loading on changes of compressive strength, thermal conductivity and structure of composites was investigated.

### 3. RESULTS AND DISCUSSION

#### 3.1 Influence of different portion of hemp hurds

Developing the compressive strength of hardened composites based on variable contents of hemp hurds after 28 and 90 days is shown in Figures 1 and 2. The highest compressive strengths reached composites containing 20 vol. % hemp hurds (the mixtures I.1 and II.1) after 28 and 90 days of hardening. In accordance with paper (Bentchikou et al., 2012), the results revealed the reduction in the compressive strength values of composite with the increase of volume portion of filler and reduction of cementitious matrix volume proportion in composites (Fig. 1 and 2). Hardened specimens examined in this study showed higher values of strength parameter than composites based on lime binder prepared under the same conditions (Kidalova et al, 2011a).

![Fig. 1. Compressive strength of composites with different hemp hurds content (mixtures I.1-I.3) after 28 and 90 days of hardening](image1)

![Fig. 2. Compressive strength of composites with different hemp hurds content (mixtures II.1-II.3) after 28 and 90 days of hardening](image2)

The same trend is observed for density values of hardened composites (Fig. 3) vs. volume percentage of hemp shives.
Higher hemp hurds share in inorganic matrix leads to lower density values of composite. The achieved densities ranged from 570 - 1700 kg.m\(^{-3}\) what are relatively low values compared with concrete but fair value for the filling elements compared to brick structures (Priganc, 2003). Linear dependence between compressive strength (R\(c\)) and MgO-cement binder ratio (c\(\text{MgO}\)) of 90 days hardened composites was obtained (R\(c\) = 0.226 \(c\text{MgO}\) – 2.551; correlation coefficient R = 0.995).

One of the most important heat technical properties of building materials is the thermal conductivity coefficient. As it can be seen in Table 3, there is observed decrease in thermal conductivity coefficient with increasing the portion of hemp hurds in the composite. Similar behaviour of composites based on different kinds of vegetable fibres was reported in papers (Khedari et al, 2001). Increasing hemp hurds share induces more porous composite what leads to the lower value of density and thermal conductivity coefficient of material.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Thermal conductivity coefficient [W/m.K]</th>
<th>Water absorbability [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>I.1</td>
<td>0.205</td>
<td>13.44</td>
</tr>
<tr>
<td>I.2</td>
<td>0.111</td>
<td>21.38</td>
</tr>
<tr>
<td>I.3</td>
<td>0.070</td>
<td>30.12</td>
</tr>
</tbody>
</table>

The porosity of the composite is determined by packing of hemp hurds slices and voids formation. Increase in water absorbability of composites with increasing portion of hemp hurds is evident from Table 3. Similar result is published in paper (Madsen et al, 2012), where the moisture content of hemp composites with a thermoplastic matrix increases linearly as a function of the fibre weight fraction.

### 3.2 Impact of mean particle length of hemp hurds

The results of measurement of physical (density, thermal conductivity coefficient, water absorption) and mechanical properties (compressive strength) of composite samples after 28 days of hardening are given in Table 4 and in Figure 4. As it is shown in Figure 4, compressive strength of prepared lightweight composites from hemp hurds depends on mean particle size of used filler material.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Density [kg.m(^{-3})]</th>
<th>Thermal conductivity coefficient [W/m.K]</th>
<th>Water absorbability [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1040±5</td>
<td>0.111</td>
<td>21.38</td>
</tr>
<tr>
<td>2</td>
<td>1070±5</td>
<td>0.115</td>
<td>25.81</td>
</tr>
<tr>
<td>3</td>
<td>1150±5</td>
<td>0.110</td>
<td>11.88</td>
</tr>
</tbody>
</table>

Compressive strength of composites increases with decreasing mean particle length of hemp hurds. The higher strength parameter value of composite with smaller mean particle length is probably due to creating a stronger structure with smaller pores, which also should be lead to higher values of thermal conductivity coefficient. The hemp particle length also appeared to influence the physical properties of composites (Stevulova et al., 2012b). The lowest value of water absorbability was recorded for composite sample 3 based on fraction with short length of hemp hurds slices. This result is connected with the densest arrangement of hemp hurds particles in MgO-cement matrix in composite. Difference in water sorption between samples 1 and 2 is caused probably by slices size in hemp hurds. Sample 2 does not content short hemp slices (fraction 8-4 mm) and structure of composite is more porous than that in the composite 1 (with a wide distribution of slices size) what led to higher value of absorbability. Whereas
the values of the water absorbability decrease in the dependence on decreasing mean particle size of hemp hurds slices, the effect of mean length of hemp hurds slices on thermal conductivity coefficient was not confirmed. This fact can be likely caused by non-homogeneous dispersion of hemp hurds slices in the matrix.

3.3 Effect of thermal loading on composites
The results of measurements of dimensions, weight and density of composites after heat treatment show a significant effect of increasing temperature on the followed parameters. As it can be seen in Table 5, the values of individual characteristics decrease with increasing temperature.

<table>
<thead>
<tr>
<th>Temperature [°C]</th>
<th>AD [mm]</th>
<th>W [kg]</th>
<th>D [kg.m⁻³]</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>99.99</td>
<td>0.987</td>
<td>990</td>
</tr>
<tr>
<td>50</td>
<td>99.86</td>
<td>0.990</td>
<td>995</td>
</tr>
<tr>
<td>100</td>
<td>98.26</td>
<td>0.871</td>
<td>920</td>
</tr>
<tr>
<td>200</td>
<td>97.23</td>
<td>0.785</td>
<td>885</td>
</tr>
</tbody>
</table>

Fig. 5 illustrated the gradual decrease in the values of the composite strength measured after heating. Composites exposed to a loading of 50°C and 100°C did not show significant decrease in compressive strength (13%). The most striking decrease in the maximum compressive strength of thermally non-treated composite was recorded in temperature range of 100°C – 200°C. The lowest value of strength parameter, which accounted for only 19.3 % of the value determined prior to thermal loading of composite, composite treated at 200°C achieves. Figure 6b shows damage of the cube sample after heat treatment at a temperature above that. Based on these results, the testing of composite confirmed their thermal stability up to 200°C.

In accordance with the work (Varga et al., 2009) a significant impact on the physico-mechanical properties of composites in addition to the temperature has duration of heat exposure, the heating rate and the possibility of exchange of moisture between the composite and the surrounding environment.

Even after heating to 200°C the coherence of composite material was maintained, which is also confirmed by the resistance to elevated temperatures. This finding is important in relation to the use of tested composites in civil construction because one of the basic requirements is resistance to high temperatures and resistance to fire, respectively.

Fig. 6. Appearance of the sample heat treated at 200°C before (a) and after strength testing (b)

Figure 7 shows morphology of original hemp shives (with magnification 80x) compared to the separated hemp slices of the composite material which were heated to 200° C.

It can be seen that the surface layers of shives are already partially thermally degraded, but mulch is still compact. This is consistent with the results published in paper (Geffert and Geffertova, 2011) which indicates the presence of lipophilic compounds.
on the surface of hemp. These organic constituents are degraded at lower temperatures due to oxidation process. The results of thermal behavior of composites based on hemp hurds slices were confirmed by FT IR study and TG measurements (Kidalova et al, 2011d; Stevulova et al, 2011). Thermal decomposition of cellulose and lignin beginning approximately at 190°C and finishing at 350°C with top at 280°C.

Values of the thermal conductivity coefficient of the thermally treated composites ranged from 0.066 to 0.099 Wm⁻¹.K⁻¹. Change in thermal conductivity coefficient in dependence on density of hardened composites before and after thermal treatment is shown in Figure 8. These values are comparable with the thermal conductivity coefficient of other building insulating materials.

![Fig. 7. SEM micrographs of original (a) and separated (b) hemp hurds slice from cube composite after heating at 200°C](image)

4. CONCLUSIONS

In this paper, the influence of different portion of hemp hurds, mean particle size of hemp shives slices and thermal loading on physical and mechanical properties of composites based on MgO-cement have been investigated. Based on results described above, following conclusions can be drawn:

- MgO-cement as alternative binder appears to be suitable for the preparation of bio-composites based on hemp hurds. Its alkaline component has a beneficial effect on the surface properties of hemp hurds.
- As the hemp hurds portion increases, values of density, compressive strength and thermal conductivity coefficient of hardened composites decrease and water absorption is being oppositely.
- Hemp hurds particles length influences the physical and mechanical properties of composites. Compressive strength and water sorption of composites after 28 days of hardening increase with decreasing mean particle length, while this effect on thermal conductivity of composites was not confirmed.
- Study of thermal loading of composites revealed that composites are damaged by increasing temperature. The most significant decrease in density and compressive strength of composites was obtained at temperature of 200°C.
- As it may be concluded from this study, hemp hurds can be used for the construction of non-load bearing walls, partitions and insulation. Products are environmentally friendly lightweight composite. However, there is still a need to study the properties of binder/matrix interface to develop durable bio-composites.

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