The study of capillarity properties of bio-composites

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Abstract. Composite materials demanded in various technical areas as well as civil engineering, have important influence on the progress in research and development of materials. Among other things, composites materials based on organic fibers, obtained from the processing of natural raw or as a product of recycling, have come to the fore in the last years. The results of the comparative study of the physical parameters (density, thermal conductivity coefficient and water absorption behaviour) of the bio-based composite are presented after 28 days of setting and hardening. The ability of the material to receive and retain water in the pore system is also very important, so parameters such as absorption and capillary absorption are also studied. Results are discussed in terms of impact of individual content of cellulotic fibers in each other properties of composite materials. Density results showed an improvement in the values with the increasing weight % fiber in composites for both types of fiber. Also, an improvement in thermal conductivity has been demonstrated, confirming the possibility of applying composites to the insulating elements. However, the results of absorption and capillary absorption imply the need to optimize the technology of preparation and surface treatment of composites.

1. Introduction

The increased quality requirements in the construction industry are also associated with the requirements for new materials. Every year, thus it is expanding the range of products that are designed to meet the requirements of consumers. In the current civil engineering, efforts are also being made to use non-traditional materials, or attention is drawn to renewable sources as hemp, cotton, line, coconut fibers etc.. Renewal of scientific as well as industrial interests in the use of cellulotic fibers as reinforcing in lightweight composite materials relates to a need of progress of environmental friendly products with high use value in term of sustainable development [1]. Due to low density, biodegradibility, interesting thermal, mechanical, acoustic and other properties of natural fibers, low cost and eco-friendly raw material, this natural fibrous material is used as a (partial) replacement of synthetic fibers, such as glass, carbon or metallic fibers [2]. The applying of natural fibers in cement based composites is narrowly linked to the ecological building sector, where a choice of materials is based on components including recyclable, renewable raw materials and low-resource manufacture [3]. Application of cellulotic fibers into cement composites must be thoroughly studied, because as it is generally known, the surface of the cellulotic materials has a hydrophilic character [4]. The hygroscopic properties expressed by absorbability and capillary water content are as basic characteristics of water transport processes into material, and recently they has been recognized as an important performance characteristic of durability materials as well [5]. This fact can be reflected both in the preparation and mixing of fresh composites, also after hardening of mixtures to their final shape. This paper is focused on specific approach to composite preparing as a way of improvement of organic filler as particulary substitution of natural sand. The aim of present study is to investigate the
utilisation of cellulosic fibers from wood pulp and waste paper as reinforcing element with insulating properties into cement composites f. e. as mortars for plastering. The major problems with the use of cellulosic fibers (both natural and recycled types) in cement composites are their high water absorption capacity, porosity and lower strength. Several research reports can be recently found worldwide, dealing with various surface treatments of fibers [2] in order to improve their surface properties. The results of the comparative study of the physical parameters (density, thermal conductivity coefficient and water absorption behaviour) of the bio-based composite are presented. Changes in capillary moisture content of composites are compared and discussed. The ability of the material to receive and retain water in the pore system is also very important, so parameters such as absorption and capillary absorption are also studied. In article these parameters were investigated after 28 days of setting and hardening. Results are discussed in terms of impact of individual content of cellulosic fibers in each other composite material. The composite durability will also be derived from the values obtained.

2. Materials and methods

2.1 Materials

The cement used in this study was Portland cement CEM I 42.5 N (Cement factory Ladce, Slovakia). Silica sand (as filler), was obtained from Šaštín - Stráže, Slovakia, (fraction size: 0/1mm). Its properties were in accordance with European standard STN EN 196-1 [7]. The two types of cellulosic fibers used in this study were - bleached wood pulp (typ GW 500), indicated in the text as sample “B” and unbleached - recycled fibers (typ G-500T), indicated in the text as sample “W” obtained from different waste papers. Both cellulosic fibers were provided by company Greencel Ltd. (Hencovce, Slovakia). Table 1 shows the basic characteristics of used cellulosic fibers. Deionised water was used in all experiments as mixing water as well as curing water. Parameters of water were in accordance with STN EN 1008 [8].

| Characteristics          | Type of cellulosic fibers |
|--------------------------|--------------------------|
|                          | GW-500 (bleached wood pulp) | G-500 T (unbleached recycled fibers) |
| Cellulose content [%]    | 99.5                     | 80                      |
| Bulk density (kg/m³)     | 60-80                    | 50-100                  |
| Max. length (µm)         | 500                      | 400                     |
| Dry matter (%)           | 93                       | 93                      |
| Ash (%)                  | 0.5                      | 0.5                     |
| pH value (-)             | 6 ± 1                    | 7 ± 1                   |

2.2. The course of experiments

The experimental cellulosic fibers reinforced composites were prepared using normal type Portland cement CEM I 42.5 N, silica sand and water. Cellulosic fibers from wood pulp and waste paper were used as particularly substitution of inorganic filler (5, 10, 15, and 20 wt. % from weight of filler). The mix proportions of specimens were designed with Cement/Sand (C/S) weight ratio of 1:3 and Water/Cement (W/C) ratio of 0.70. Reference cement composite was manufactured without using of cellulosic fibers. Cube samples of 100mm x100mm x100 mm were prepared and cured under standard conditions. All of the experimental mixtures were obtained in a laboratory by using a laboratory mixer.

The density, coefficient of thermal conductivity of composites with various content of cellulosic fibers (5, 10, 15 and 20 wt. %) were investigated after 28 days of hardening. Density was calculated from the weight and the determined dimensions of the individual samples.
The thermal conductivity coefficient of samples as the main parameter of heat transport $\lambda$ (W.m$^{-1}$K$^{-1}$) was measured by the commercial device ISOMET 104 (Applied Precision Ltd., Germany). The determination is based on the analysis of the temperature response of the analyzed material to heat flow impulses. The heat flow is induced by electrical heating using a resistor heater having direct thermal contact with the surface of the sample.

Determination of absorbability was performed by immersing the dried samples into a container with deionised water at 20 ±2°C. The evaluation of absorbability was carried out gradually for 1, 6, 12 and 24 hours, when the pores of the composites were filled completely with water. Total water absorbability of composites is represented in table 3 (evaluation after 24 hours testing). This way is in contrast to the determination of total water absorbability (when samples are immersed completely into water bath).

In addition, the capillary water content was tested after 28 days of hardening. Samples (cubic type 100mm x 100mm x 100mm) were dried up to constant mass. In case of capillarity determination, samples are immersed in water at a depth of 5–10 mm for a specific period of time only by defined area of sample (exactly specified in mm$^2$). The test procedure consists of recording, at defined time intervals, the mass increment by capillary suction of a specimen with contact surface of samples with water, up to constant weight difference between two consecutive weightings less than 0.1%.

For this experiment, measurements were done in one step in the following intervals: 300s, 600s, 900s, up to 3600s, consequently at next measurements to show the development of this parameter in time up to 7 days. All procedures were performed in accordance with standards rules [9], [10]. The values of the coefficient of the capillary water absorption were calculated by the equation (1):

$$ c = \frac{(m_{ti} - m_{t0})}{A_s} $$

where:
- $c$ - coefficient of the capillary water absorption
- $m_{ti}$ - specimen mass after the process of soaking (g) in time $t = i$ (s)
- $m_{t0}$ - specimen mass after the process of drying (g) in time $t = 0$ (s)
- $A_s$ - cross area of the specimen surface immersed in water (m$^2$)

3. Discussion of results

3.1. Density and thermal conductivity of fibers cement composites

Table 2 reports the values of important parameters such as density and thermal conductivity of fiber cement composites as well as reference composite. While density of reference sample after 28 days of hardening reached 2110 kg/m$^3$, specimens with cellulosic fibers GW-500 (sample B) in an amount of 0.5 wt. % to 20 wt. % as a filler substitute, showed the decrease in density 6.16% up to 8.53% compared to the reference sample. The lowest values of density composites were determined in case of composites on the base G500-T (unbleached recycled fibers) (sample W). There was observed reduction in density of composites from 3.32% up to 4.98% compared to the reference sample (see table 2). Reduction in density values is a positive fact, it confirms the possibility of composites using as insulating element of constructions. To reduce the density of composite has also technological importance, because it can improve the workability of fresh mixture, when using composite as mortar for construction.

Regarding the evaluation of thermal conductivity values it can be stated that there is in all the samples a decrease in average values compared to the reference sample. The lowest value of thermal conductivity (0.291W/m.K) and its maximal reduction (30.38 %) of 28 days hardened fiber cement composites was found in case of the sample 20% W in comparison with reference sample (0.418 W/m.K). The remaining detail results of samples "W" and their comparison with reference sample are in table 2. Reducing the coefficient of thermal conductivity with the increasing content of cellulose
fibers was also observed in samples based on bleached cellulose samples (B), however this reduction, compared with reference sample was 12.44% (sample 5%B) in case sample 20%B% it was 21.77%. Generally, the fiber additions into matrix have positive impact on density and thermal conductivity of fibers cement composites, as well. Incorporation of cellulosic fibers into structure of composites creates the voids and the heat-insulating properties are enhanced when density decreases.

Table 2. Density and thermal conductivity values of cement composites.

| Sample   | Density $\pm 5$ (kg.m$^{-3}$) | Change (%) decrease of values | Coefficient of thermal conductivity $\pm 0.001$ (W/m.K) | Change (%) decrease of value $\lambda$ |
|----------|--------------------------------|------------------------------|--------------------------------------------------------|-------------------------------------|
| Reference| 2110                           | 100/0                        | 0.418                                                  | 100/0                               |
| 5%B      | 1980                           | 93.84/-6.16                  | 0.366                                                  | 87.56/-12.44                        |
| 10%B     | 1945                           | 92.18/-7.82                  | 0.361                                                  | 86.36/-13.64                        |
| 15%B     | 1935                           | 91.71/-8.29                  | 0.358                                                  | 85.64/-14.36                        |
| 20%B     | 1930                           | 91.47/-8.53                  | 0.327                                                  | 78.23/-21.77                        |
| 5%W      | 2040                           | 96.68/-3.32                  | 0.350                                                  | 83.73/-16.27                        |
| 10%W     | 2030                           | 96.20/-3.80                  | 0.332                                                  | 79.43/-20.57                        |
| 15%W     | 2015                           | 95.49/-4.51                  | 0.304                                                  | 72.73/-27.27                        |
| 20%W     | 2005                           | 95.02/-4.98                  | 0.291                                                  | 69.62/-30.38                        |

3.2 Assessments of hygroscopic properties of composites

Generally, the assessments of the hygroscopic properties of building elements - composites are of considerable importance also from the service life point of view [6]. They also relate to parameters such as frost resistance or durability, and hence the location and method of composites application in building constructions. From hygroscopic parameters has focused attention on assessment of water absorbability of composites, being for technologically application of composites is important, whether it is the total water absorption or capillarity.

3.2.1 Determination of absorbability. The results of total absorbability show the expected increasing of amount of water in the composites, both over time and with the increasing content of cellulose fibers as well (table 3). Increasing the water absorbability also may be caused by rising of fiber amount in cement matrix because cellulosic fibers have hydrophilic character [1].

3.2.2 Capillary absorption testing of composites. The data on the capillarity properties obtained from all of the produced composites (with 5%, 10%, 15% and 20% GW-500 fibers mixes (B), and 5%, 10%, 15% and 20% of G-500T fibers mixes (W) are shown graphically. The results of the determination of capillary absorption at the time up to 1 hour for individual mixes composites (W) are shown in figure 1 and composites (B) in figure 2. A comparison of the development capillary absorption results shows, that composites (B) with a lower % fiber replacement of filler exhibit lower values over the reference sample except sample 20% B (with density 2005 kg/m$^3$).

Regarding the results of the capillary absorption for the composites (W), see figure 1, that a growing share of fibers as a substitute filler occurred seal the pores, thereby affecting the ability of soaking. This knowledge is very important in terms of the intended use of composites as a plaster. In further experiments, long-term capillary absorption also will be tested from the point of view of the durability of the composites.
Table 3. Water absorbability of composites after 24 hours testing.

| Sample | Absorbability ±0.01 % | % changes |
|--------|------------------------|-----------|
| Reference | 8.85 | 0 |
| 5% B | 9.85 | +11.29 |
| 10% B | 11.21 | +26.66 |
| 15% B | 11.24 | +27.05 |
| 20% B | 11.93 | +34.80 |
| 5% W | 10.01 | +13.10 |
| 10% W | 10.36 | +17.06 |
| 15% W | 10.42 | +17.18 |
| 20% W | 10.83 | +22.37 |

Figure 1. Results of the capillary absorption test (1 hour) mixes of composites (W).

Figure 2. Results of capillary absorption test (1 hour), mixes of composites (B).
4. Conclusion
The quality and durability of composites is strongly dependent on the kind, properties and dosage of the basic components. The cellulosic fibers due to their low density, thermal, mechanical and other technical properties are used as a partial replacement of inorganic filler or another synthetic fiber into cement composites. It is clear, that the technological procedures of composites preparing or the way of coating material can influenced the terminal properties of products from durability point of view. The capillary water content as well as absorbability are important parameters which must be take in count, because of increasing water content in construction threatens to create disorder, failures, up to an emergency.

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