Wood pulp as a potential raw material source for manufacturing bio-based building materials

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Abstract The application of bio-based material such as cellulose fibres derived from renewable source to cement mortars was investigated and the properties of fresh mixtures and hardened mortars were characterized. The mixtures composition consisted of combined addition of the selected kind of cellulose fibres (bleached wood pulp) and superplasticizer (polycarboxylate ester). The aim was to produce a cement mortar with the improved properties for indoor application. The 0.5 wt.% fibre addition (from weight of filler and binder) and superplasticizer dosage of 1% in the mixtures were used. Reference cement mortar did not contain cellulose fibre and superplasticizer. The comparative study of the fresh fibre cement mixture properties has been showed worse flowability behaviour of mixture with plasticizer, as expected. The beneficial effect of the superplasticizer was not proven even on the compressive and flexural strength of fibre cement mortars after 7, 28 and 90 days of hardening. The performances of the mortars were dependent not only on fibres and superplasticizer nature but also on the organic/inorganic interface.

1. Introduction
Nowadays, the use of raw material from renewable sources to bio-based building materials is significantly interested in terms of development and production of environmentally friendly materials. Generally, renewable sources are considered as option in their use in building materials to improve the sustainability of the construction industry [1]. To improve sustainability in building materials usage, the construction industry must include the reuse of industrial by-products and renewable materials in construction [2]. Natural fibres in cement composites are becoming increasingly popular. The use of cellulosic fibres as a renewable source coming from different sources such as wood, vegetables, agro-plants and waste from their processing in the production of cement composites has drawn an increasing interest [3, 4]. Cellulosic fibres offer a variety of advantages such as wide availability, biodegradability, fast recoverability at relatively low costs, low density, adequate stiffness and strength, variety of morphologies, controllable aspect ratio and surface roughness, an interfacial compatibility through appropriate modification of the fibre surface and are without known health hazards [5]. Natural fibres obtained from renewable sources are readily available and a relatively cheap material compared to man-made fibres. The utilization of cellulose fibres into cement-based composites/mortars in European today’s construction industry is renewed since the last century due to environmental benefit, energy and resource conservation [2].
The incorporation of different type and size of cellulose fibres into cementitious matrix led to favourable effects regarding the reduction of matrix brittleness and increase in the durability, which is proportional to the resistance to crack propagation offered by the fibres bridging the matrix, thereby effectively transferring the load [6]. Many research works on the use of cellulose fibres coming from different renewable sources such as wood [7, 8, 9], sisal [10, 11], hemp [12-15], coconut [11], and bamboo [16, 17] in bio-based fibre cement materials have been performed and reported. The use of cellulose fibres and fibrous waste residues promotes bio-architecture and relates to low environmental impact at the end of the lifecycle.

In the present work, the cellulotic fibres coming from wood pulp as a potential source of raw material are incorporated to the cement mixture. This study is focused on the properties of fresh and hardened cement mortars containing wood pulp in dosage of 0.5 % (respect to binder and filler weight) and superplasticizer addition (1 % from cement weight). The flowability of fresh mixture, density (28 days hardening time) as well as mechanical properties (compressive and flexural strength) of hardened fibre cement composites after 7, 28 and 90 days of hardening were studied.

### 2. Materials and methods

#### 2.1. Materials

CEM I 42.5 N, the ordinary Portland cement supplied by Cement Factory Ltd. (Povazska cementarna Ladce, Slovakia) was used for the preparation of the cement mortars. Standard natural silica sand from company Filtracni pisky Ltd (Chlum, Czech Republic) in accordance with standard STN EN 1008-1 [18] was used as filler. The wood pulp Greencel was obtained from Slovak company Greencel Ltd (Hencovce, Slovakia). The white bleached wood pulp GW-500 with 99.11 % of holocellulose content was used as 0.5 % addition in relation to the total weight of the filler and binder. The physical properties of this sample of cellulose fibres are shown in Table 1. Water for the cement mixtures preparation was used in accordance with standard STN EN 1008 [19]. To adjust the workability of fibre cement mixtures a superplasticizer MasterGlenium Sky 665 (as polycarboxylate ester type) acquired by BASF - Chemical Company Ltd (Prague, Czech Republic) was taken in dosage of 1 % from weight of cement according to standard STN EN 934-2+A1 [20].

| Aspect ratio | Thermal conductivity [W/m.K] |
|--------------|----------------------------|
| 23.2         | 0.0674                     |

#### 2.2. Specimens preparation, curing end testing

Three different formulations were used for the preparation of the mortar samples. The weight ratio of components of cement: sand: water was kept 1: 3: 0.55 in the mix design. Mixture sample WPA contained wood pulp addition of 0.5 wt. % from weight of cement and sand. Unlike this sample, the WPAP mixture was also prepared with the addition of a plasticizer of 1 wt. % dosage from weight of cement. For comparison to mixtures WPA and WPAP, reference sample (RF) without wood pulp as well as superplasticizer was prepared.

Preparation of mortars started with soaking of wood pulp and manual mixing in approximately 50 wt. % of water. Remaining water, superplasticizer (only for WPAP sample), the required amount of sand and the cement was mixed by mechanical stirring in a mixer in accordance with standard [18] (to ensure the homogenous distribution of wood pulp in cement mixture). Mortars bodies were produced by placing the fresh mix to standard steel prism in the dimension of 40 x 40 x 160 mm. The samples of mixture WPA were consolidated using a jolting apparatus and for the samples WPAP (prepared with
wood pulp and superplasticizer) were used a vibrating table. Specimens were demoulded one day after mixing and subjected to the following curing regime. Specimens were kept in water under laboratory temperature (+ 20 °C) for 7, 28 and 90 days.

Flow behaviour of the fresh fibre cement mixtures (indicating its workability) was estimated by a flow table test in accordance with standard STN EN 1015-3 [21].

Dry bulk densities of fibre cement mortars were calculated after 28 days of hardening [22].

The thermal properties such as thermal conductivity, thermal diffusivity and volume heat capacity were experimentally determined. Before testing these parameters, all specimens (40x40x160 mm) were dried at 105°C until a constant mass is reached. The thermal parameters were measured by using an ISOMET 2114 with a planar sounder.

The strength tests of 7, 28 and 90 days hardened fibre cement composites were carried out by using a compression test machine (FORM+TEST Seidner & Co. GmbH, Riedlingen, Germany) according to standard [23]. In the case of the compressive strength testing, a loading rate was 2 400±200 N/s, while the three-point bending test was performed at a loading rate 50±10 N/s. To determine the fibre cement mortar physical and mechanical average values, series of three prisms with and without fibres were fabricated.

3. Results and discussion

3.1. Consistency of fresh mixtures

Consistency measurements of three of fresh wood pulp cement mortar samples with constant water/cement ratio of 0.55 are illustrated in Figure 1. Test results of their flow ability are presented in Figure 2. As known, the flow diameter value of the fibre mixture is proportional to the fluidity of the matrix. In the papers [24, 25], there was noticed the influence of cellulosic fibres on flow ability of fresh cement mixture. In other study [26], the superplasticizer was used for better flow ability as well as improvement the workability of fibre reinforced cement mortars and it is generally preferable for moldability and practical use. Polycarboxylate-based superplasticizers significantly affect the properties of fresh and hardened concrete/mortar by their physical and chemical properties. Polycarboxylates are formed by long side chain organic macromolecules that adsorb on the surface of the cement particles and due to the steric effect significantly affect the fresh mortars consistency. This leads to their dispersion, deflocculation and the stabilization of the cement mixture. This creates better conditions for hydration and for increasing the strength of cement composites.

![Figure 1. Consistency of reference sample RF (a) and fiber cement fresh mixtures WPA (b) and WPAP (c) with addition of cellulose fibers (0.5 wt.%) and superplasticizer (1 wt.%).]

Our experiments demonstrated that the flow values decreased with incorporation of cellulosic fibres and superplasticizer addition into fresh cement mixture in comparison to cement mixture RF.
Figure 2: Flow diameter values of fresh fibre cement mixtures.

(without fibres and superplasticizer). This decrease was 51.5 % and 43.9 % for WPA and WPAP, respectively. WPA sample had the lowest value of flow diameter (116 mm). The reason for this behaviour of fresh fibre cement mixtures (without plasticizer) is that the natural fibres have a hydrophilic nature, causing absorption of larger amounts of water and it consequently significantly reduces the resulting consistency of the fresh mixture. WPAP sample with superplasticizer reached a slightly higher value (134 mm) than WPA mortar. The goal of using the superplasticizer into fibre cement mixture was to achieve or approach the consistency value of the reference sample. The experiment showed that the selected superplasticizer used in the given amount did not improve consistency of fibre cement mixture.

As known, the main factors affecting the rate of workability loss of fresh concrete are the initial consistency, the type and amount of plasticizer, the type and amount of cement, the time of addition of the admixture, the laboratory conditions (temperature, humidity), the mixing method and the presence of other additives [27]. Authors assume that one of the main factors why the consistency of the reference sample is not achieved is the presence of organic fibers. Based on results of the flow behaviour study of recycled paper cellulosic fibre cement composites [28], the surface nature of fibres is the important factor determining the consistency of fresh fibre cement mixture.

3.2. Density and thermal conductivity of fibre cement based mortars
The density results of 28 days hardened mortars are shown in Figure 3. In accordance with published work [29], the addition of lightweight porous fibres occupies part of the volume of cement specimen resulting in a density reduction.

The decrease in density of the fibre cement mortar is recorded due to lower density of cellulosic fibres than density of cement [30]. Reduction in density is also caused by fibre incorporating air into the matrix during the mortar mixing. However, this occurrence of this phenomenon becomes more significant in the case of using high percentages of fibres [31]. The WPA mixture with a small amount of wood pulp (0.5 wt. %) led to relatively lower density value by 4% as compared to RF sample but on the other hand WPAP fibre cement sample with addition of superplasticizer showed very slightly gain in density (1.2 %) in comparison to RF sample. These differences in density values are at the measurement error level.

The average values of thermal conductivity, thermal diffusivity and volume heat capacity of 28 days hardened fibre cement mortar samples are given in Table 2. The values of thermal conductivity and thermal diffusivity are in line with the trend of density values. Whereas the decrease in the thermal
conductivity was 20% for sample WPA compared to RF sample, higher value of thermal conductivity coefficient was measured for WPAP sample than for WPA mortar sample. As shown in [32], a higher content of cellulose fibres in the mortars (4-12%) led to a lower thermal conductivity values, i.e. a greater thermal insulation capacity.

Table 2. Thermal conductivity, thermal diffusivity and volume heat capacity for 28 days hardened fibre cement mortars and reference sample.

| Sample | Thermal conductivity $\lambda$ [W/m.K] | Thermal diffusivity $\alpha \times 10^6$ [m$^2$/s] | Volume heat capacity $c_p \times 10^6$ [J/m$^3$.K] |
|--------|--------------------------------------|---------------------------------|----------------------------------|
| RF     | 2.699                                | 1.607                           | 1.679                            |
| WPA    | 2.162                                | 1.273                           | 1.697                            |
| WPAP   | 2.45                                 | 1.299                           | 1.885                            |

3.3. Mechanical properties of fibre cement based mortars

The development of compressive and flexural strengths on hardening time (7-90 days) is illustrated in Figures 4 and 5. According to the experimental results, the strength values have been reduced by incorporating of the fibre content to the cement matrix. The WPA cement mixtures containing only wood pulp reached lower values of the compressive strength by 16.6 % in comparison to reference sample after hardening time of 90 days. The compressive strength values of fibre cement mortars with a superplasticizer addition (WPAP mortar) were very close to the RF sample in each hardening time. The compressive strength values of 28 day hardened fibre mortars are in the accepted range for plastering mortars [33].

As shown in Figure 4, the addition of cellulose fibers did not enhance the flexural strength of the fibre cement specimens (WPA sample) in the prolonged hardening time, the dosage of superplasticizer to the fibre cement mixture (WPAP sample) caused improvement in the tested strength parameter as compared to WPA mortars. These findings are in accordance with the published research results where a general reduction in mechanical properties of fibre reinforced cement composites was observed [34, 35]. This decrease in strength parameters of fibre cement samples is attributed to the creation of air voids due to the incorporation of porous cellulose fibres in the cement matrix resulting in a reduction of bond strength of the matrix, causing overall reduction in strength [36]. As generally known, the application of a superplasticizer admixture in the cement mixture can improve workability, increase
strength, conserve cement and help reduce shrinkage and thermal cracking due to maintains the fluidity and providing the mix stability.

Figure 4. Compressive strength of hardened fibre cement mortars after 7, 28 and 90 days.

Figure 5. Flexural strength of hardened fibre cement mortars after 7, 28 and 90 days.

As reported above, the performance of the fresh fibre cement mixtures was not improved by a small addition of superplasticizer. The plasticizer efficiency control parameter is strength of hardened mortars. The beneficial effect of polycarboxylate superplasticizer on the compressive and flexural strengths of WPAP fibre cement mortar specimens at all hardening times has been not recorded, although the works [37, 38] demonstrated that the presence of a superplasticizer in the cement matrix has a favourable impact on the compactness of the structure; plasticizer reduces the pore size or numbers the pores and as a result the compressive strengths are increased. However, the introduction
of fibre into the cement mixture did also not promote a significant increase in the strength of the mortars both in compression and in bend. It is likely that the cause of this reduction in mechanical properties of fibre cement based mortars is the presence of cellulosic fibers and their surface properties. The properties at the cement particles/cellulosic fibres interface play a key role in the mortars toughness [16]. The hardened cement specimens based on waste paper cellulosic fibres with lower holocellulose content and the addition of same superplasticizer achieved the highest values of compressive (48.4 MPa) and flexural (up to 7 MPa) strength due to improved bond strength between fibres and matrix [28].

4. Conclusions
Two fibre cement mortars containing cellulose fibres from bleached wood pulp in 0.5 wt.% addition (from weight of filler and binder) and superplasticizer based on polycarboxylate ester (dosage of 1% of cement weight) were prepared. The objective of this study was to determine the effect of superplasticizer addition on the workability of fibre fresh mixtures and on the physical and mechanical properties of hardened specimens in comparison to the parameters for reference sample. The results can be concluded as follows:

- the minimum differences in density values of fibre cement mortars with and without superplasticizer in comparison to reference sample were obtained. The variance of calculated density values was at the measurement error level.
- the flow values of fresh cement mortars, the thermal and mechanical properties (compressive and flexural strength) of hardened mortars were dependent on the presence of cellulosic fibres and superplasticizer in mixtures compared to reference cement mixture (without fibres and superplasticizer).
- the performances of the mortars at all hardening times were dependent not only fibres and superplasticizer nature but also on the organic/inorganic interface.
- the values of compressive strength of 28 days of hardened fibre cement mortars are in the accepted range for plastering mortars.

Preliminary investigation of the impact of superplasticizer on the properties of fresh and hardened fibre cement mortars under the studied conditions revealed that the selected superplasticizer used in the given amount is not appropriate for fibre cement mixture. Further research will be therefore focused on the selection plasticizer of another material base and optimization of cellulosic fiber cement mortar composition to develop thermal insulating cement mortar.

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