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Recycled Cellulosic Fibers and Lignocellulosic Aggregates for Sustainable Building Materials

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***Abstract*—**Sustainability is becoming a priority for developers and the use of environmentally friendly materials is increasing. Nowadays, the application of raw materials from renewable sources to building materials has gained a significant interest in this research area. Lignocellulosic aggregates and cellulosic fibers are coming from many different sources such as wood, plants and waste. They are promising alternative materials to replace synthetic, glass and asbestos fibers as reinforcement in inorganic matrix of composites. Natural fibers are renewable resources so their cost is relatively low in comparison to synthetic fibers. With the consideration of environmental consciousness, natural fibers are biodegradable so their using can reduce CO_2 emissions in the building materials production. The use of cellulosic fibers in cementitious matrices have gained importance because they make the composites lighter at high fiber content, they have comparable cost - performance ratios to similar building materials and they could be processed from waste paper, thus expanding the opportunities for waste utilization in cementitious materials. The main objective of this work is to find out the possibility of using different wastes: hemp hurds as waste of hemp stem processing and recycled fibers obtained from waste paper for making cement composite products such as mortars based on cellulose fibers. This material was made of cement mortar containing organic filler based on hemp hurds and recycled waste paper. In addition, the effects of fibers and their contents on some selected physical and mechanical properties of the fiber-cement plaster composites have been investigated. In this research organic material have used to mortars as 2.0, 5.0 and 10.0 % replacement of cement weight. Reference sample is made for comparison of physical and mechanical properties of cement composites based on recycled cellulosic fibers and lignocellulosic aggregates. The prepared specimens were tested after 28 days of curing in order to investigate density, compressive strength and water absorbability. Scanning Electron Microscopy examination was also carried out.

***Keywords*—**Hemp hurds, organic filler, recycled paper, sustainable building materials.

I. Introduction

THE construction industry is one of the major and most active sectors in Europe. It represents 28% and 7% of employment respectively, of the industry and of all the European economy. Unfortunately, this industry is also responsible for the depletion of large amounts of non-renewable resources and for 30% of carbon dioxide gas emissions [1]. The use of renewable resources by the construction industry will help to achieve a more sustainable consumption pattern of building materials as well as using planning tools for sustainability [2], [3].

In the last few years, an increase in interest has been given to the use of cellulose fibers and lignocellulosic biomass as alternatives for conventional reinforcements in composite materials for sustainable building construction [4]. The development of commercially viable environmentally friendly and healthy materials based on natural resources is on the rise. In this sense, cellulosic fibers as reinforcements for cement mortars constitute a very interesting option for the construction industry [5].

Biocomposites based on cellulosic fibers with inorganic matrix represent a group of lightweight materials providing a healthy living in buildings [6]. Over the last few years, problems related to environmental issues have motivated extensive research on environmentally friendly materials. Particular interest has been given to the use of fibers obtained from renewable vegetable sources in composite materials [7]. A combination of interesting mechanical and physical properties and their environmental benefits has been the main driver for their use as alternatives for conventional reinforcements. Vegetable or cellulose fibers exhibit a set of important advantages, such as wide availability at relatively low cost, biorenewability, ability to be recycled, biodegradability, non-hazardous nature, zero carbon footprint, and interesting physical and mechanical properties (low density and well-balanced stiffness, toughness and strength) [8], [9].

Nowadays, the need for sustainable, energy efficient construction materials has oriented extensive research on alternative materials to produce environmentally friendly construction products. Applications of biocomposites are basically addressed to the non-structural building of thin walled materials, mainly thin-sheet products for partitions, building envelope or ceilings flat sheets, roofing tiles and prefabricated components in general [10]. According to their origin and composition, cellulosic fibers are classified as non-wood and wood fibers. Wood fibers are also known as lignocellulosic fibers because they have higher lignin content than non-wood fibers [11].

Fiber reinforced cement composites exhibit improved toughness, ductility, flexural capacity and crack resistance compared with non-fiber reinforced cement-based materials. The major advantage of fiber reinforcement is the behavior of

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the composite after cracking has started, as the fibers bridge the matrix cracks and transfer the loads. The post cracking toughness may allow more intensive use of such composites in building. Cellulosic fibers provide adequate stiffness, strength and bonding capacity to cement-based matrices for substantial enhancement of their flexural strength, toughness and impact resistance [12], [13]. Moreover, these fibers can reduce the free plastic shrinkage [14]; decrease the thermal conductivity [15] and improve the acoustic performance increasing the sound absorption and the specific damping and the density of the composite [16].

In this study, two types of plasters based on bio-based waste (cellulosic fibers coming from recycled paper and lignocellulosic aggregates coming from hemp stem processing) were prepared and analyzed to evaluate the influence on their performance of different fibers used as reinforcement. In particular, the same amount (2%, 5% and 10%) of recycled fibers and hemp hurds (Cannabis Sativa) was adding to the mortar.

This paper reports determination morphological features of recycled cellulosic fibers and lignocellulosic aggregate using by scanning electron microscopy (SEM) and EDX investigation. Hemp hurds as lignocellulosic aggregate and recycled fibers from waste paper are introduced in mortar and their effect on physic-mechanical parameters (density, water absorbability, compressive strength) is studied in dependence on the percentage of used fibrous material.

II. Materials and Methods

A. Recycled Cellulosic Fibers

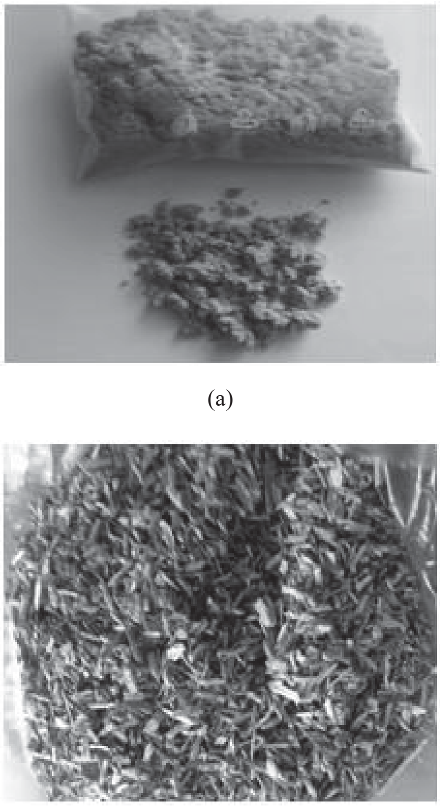
Cellulose fibers from recycled paper (Fig. 1 (a)) - Greencel were supplied by Bukoza Invest Ltd (Hencovce, Slovakia). The cellulose content of recycled paper fibers is 80% and the remaining percentage is represented the ash. The mean particle size of the cellulosic fibers is 1.2 mm and value of bulk density is 30-50 kg/m^3.

B. Lignocellulosic Aggregates

Technical hemp hurds obtained from hemp processing coming from the Netherlands Company Hempflax was used as an aggregate in the experiments. Original hemp hurds slices had wide particle length distribution (8 – 0.063 mm). For this experiment fraction <2 mm (Fig. 1 (b)) was prepared after sieving and the mean particle length of this hemp material was 0.94 mm. The measured bulk density of hemp hurds was 117.5 kg/m^3. The chemical composition of used lignocellulosic hemp material is shown in Table I.

TABLE I
CHEMICAL COMPOSITION OF HEMP HURDS

Hemp hurds component	Content (%)
Holocellulose	77.28
Cellulose	44.5
Hemicelluloses	32.78
Lignin	21.03
Components soluble in toluene-ethanol extract	3.57
Ash	3.04



(a)

(b)

Fig. 1 Organic filler (a) cellulose recycled fibers; (b) hemp hurds

C. Scanning Electron Microscopy and Energy Dispersive X-Ray Spectroscopy of Organic Filler

Scanning electron microscopy (SEM) observations and Energy-dispersive X-ray spectroscopy (EDX) characterization were done on a TESCAN MIRA 3 FE (TESCAN, Brno, Czech Republic). Characterization was done on cellulose fibers from recycled paper and hemp hurds. Fiber samples were glued on carbon adhesive films and coating with carbon film using a vacuum sputtering coater. The samples were coated with carbon film to avoid charging under the electron beam.

D. Samples Preparation and Testing

Two kinds of organic filler – recycled paper fibers and hemp hurds in various portions (2.0, 5.0 % and 10.0%) were selected for preparation of cement mortar composite samples.

Portland cement CEM I 42.5 R (Holcim Slovakia a.s.) as binder and natural silica sand (fraction 0-0.6 mm) as filler were used into mixtures. Water for the cement mixtures preparation was used in accordance with standard STN EN 1008. Fresh reference mortars were prepared with Cement/ Sand (C/S) weight ratio of 1:3 and Water/Cement (W/C) ratio of 0.75. Each batch of three test specimens consists of 450 ± 2 g of cement, 1350 ± 5 g of sand and 337 ± 1 g of water. Fresh mortar plus organic fibers were prepared with cement/(sand + fibers) weight ratio of 1:3. The fiber content was subtracted from the silica sand content which means that fibers were used as substitutions. The sand content was then adjusted based on mixing rule where the ratio water/cement is kept constant.

Preparation of fiber reinforced cement composites was carried out in two different ways. At first, the recycled paper fibers were dispersed in water by mechanical stirring (approximately 50 wt. % of water). Subsequently, cement, sand and remaining amount of water were added, and mixing continues to allow uniform fiber dispersion in the mixture. At second, the components of mixture with hemp hurds were homogenized in dry way and then mixed with water addition. After mixing, the mortars were immediately poured into the standard steel block forms with dimensions 40 x 40 x 160 mm.

The specimens were cured for 2 days in the indoor climate at approximately +18°C and then they were removed from the forms. After that time, the specimens were held under PVC foil for 26 days. For each measurement were prepared 3 samples.

The density of cement mortar composites were determined in accordance with standard STN EN 12390-7 and STN EN 12390-3.

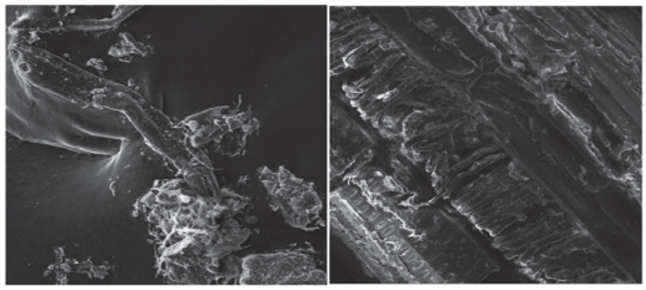
Short term water absorption (after 1h) was carried out in accordance with the standard STN EN 12087/A1.

Compressive strength of the all composite samples was being estimated after 28 days of hardening. Tests were carried out in accordance with the requirements of the standard STN EN 206-1/A1 using a laboratory instrument ADR ELE 2000 (International Limited, United Kingdom) as the maximum load per average cross-sectional area.

III. Results and Discussion

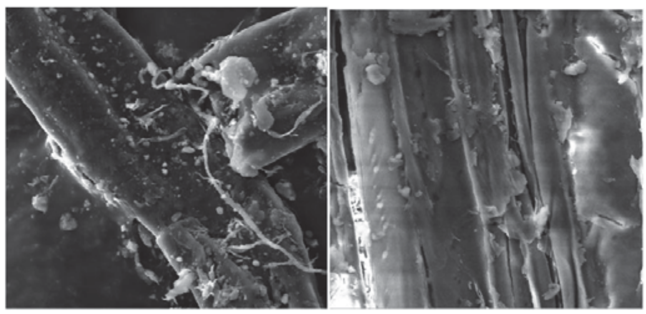
A. SEM and EDX characterization

The fiber quality was checked using Scanning Electron Microscopy (SEM) to reveal surface roughness, imperfections and overall geometry. Examinations were carried out on the recycled waste paper fibers and on hemp hurds to find out the morphological characterizations. The SEM micrographs of cellulose fibers surface (a) and of hemp hurds (b) are shown in Figs. 2 and 3. Recycled paper fibers have on their surfaces impurities that come from re-pulped waste paper (ink, different resources of waste paper and filler in paper making). Hemp hurds surface topography shows the presence of surface impurities like ash and waxes. The fiber structure is formed by several bundles of filaments aligned the plant´s length.



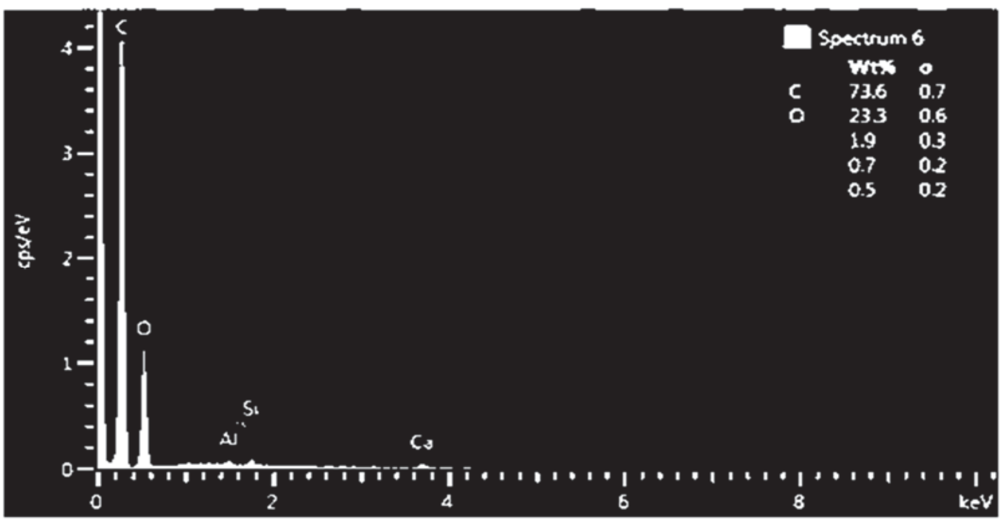
(a) (b)

Fig. 2 SEM micrographs of recycled cellulose fibers (a); hemp hurds (b) (1500 times of magnification)

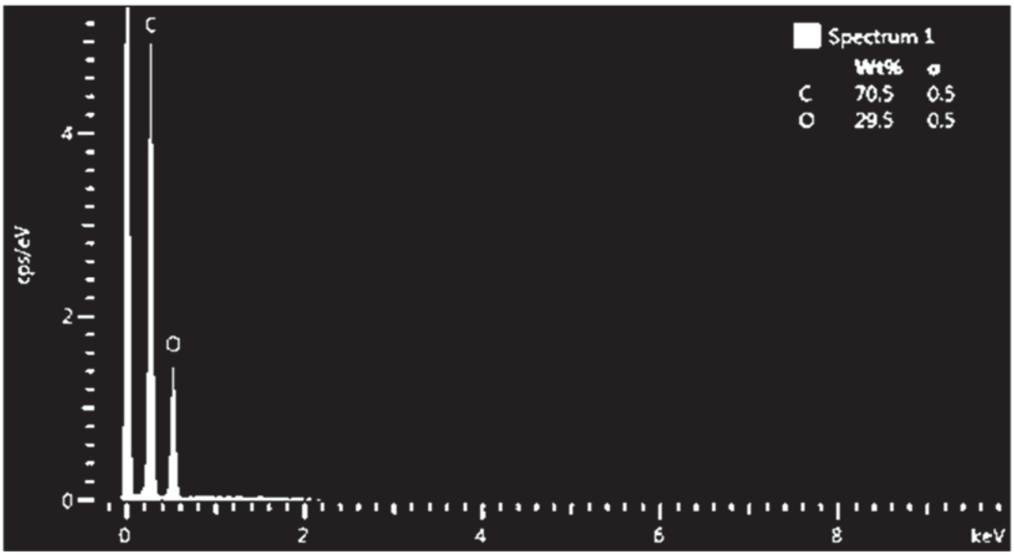


(a) (b)

Fig. 3 SEM micrographs of recycled cellulose fibers (a); hemp hurds (b) (6000 times of magnification)



(a)



(b)

Fig. 4 EDX investigation of cellulosic fibers from waste paper (a); lignocellulosic hemp hurds (b)

The chemical elements analysis of the recycled paper fibers and hemp hurds was studied by Energy-dispersive X-ray spectroscopy (EDX). The method EDX was used to identify elements present in these bio-based materials. The elements representations of recycled fibers and hemp hurds are depicted in Fig. 4. There are showed and investigated signals of C (carboneum) and O (oxygenium) which came from their native character. The EDX analysis of fiber surface coming from waste recycled paper confirmed presence signals of Al (aluminum), Si (silicium) and Ca (calcium). These inorganic elements are impurities coming from the paper processing. Whereas, the original hemp hurds has natural character there was not detected any signals of inorganic elements.

B. Physical and Mechanical Parameters of Composites

For this investigation, various mixtures of fiber-cement composites were prepared. Cellulose waste fibers and lignocellulosic hemp aggregates were added with different percentages (2 wt. %, 5 wt. % and 10% wt. replacement of sand) into cement mortar composites and a reference sample without fibers addition was prepared for comparison. The all physico-mechanical parameters were measured on composite samples after 28 days of hardening.

The first measured parameter of fiber-cement mortar samples was density. In Fig. 5, dependence of the percentage of cellulose fibers on composites density is shown. The both types of composites, cement mortar samples with recycled waste paper fibers and also samples with hemp hurds show decrease of density with increasing amount of fibers. The visible decrease of cellulosic fibers cement mortar composites density is in the range 4.44–7.72% in comparison with reference sample. Density of samples prepared with lignocellulosic hemp hurds have significantly decreased: 8.88; 10.08 and 13.31% in 2.0; 5.0 and 10.0 wt.% of filler replacement in comparison with reference sample, respectively. This is caused by amount of fibers, their nature and their physicochemical characteristics [17].

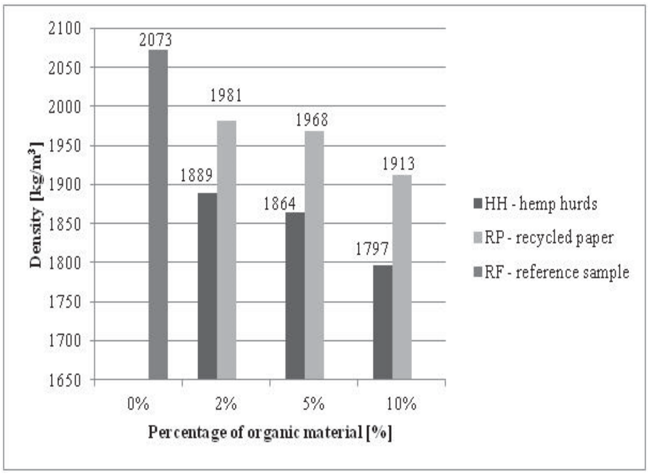


Fig. 5 Influence of organic filler content on composites density

Fig. 6 shows the mechanical behavior of two mixture types on various contents of organic filler to cement mortars in comparison with reference sample after 28 days of hardening. The compressive strength values of all mixtures are lower than value of reference specimen. The compressive strength tends to increase with increasing fiber content up to a point where there is a decrease in compressive strength of samples due to phenomenon, when an excessive amount of fibrous material reduces the strength. In this case, as it is shown in Fig. 6, there is recorded an increase of compressive strength up to 5% wt. content of waste paper fibers and hemp hurds at 2.70% and 6.33%, respectively.

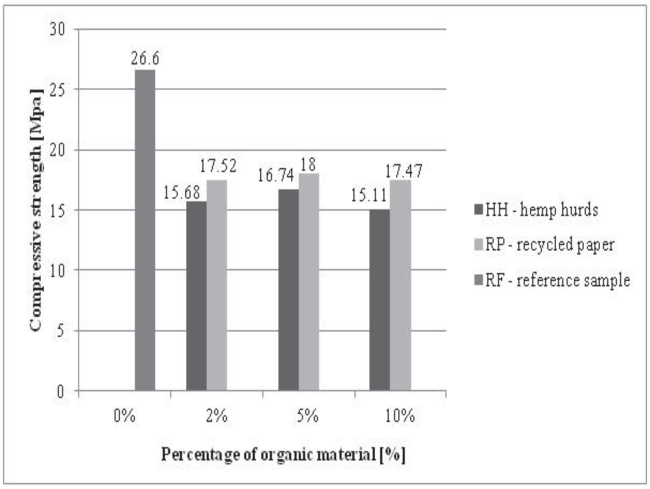


Fig. 6 Influence of organic filler content on compressive strength

The relationship between compressive strength and density values of the all composites with organic filler is depicted in Fig. 7.

In this case, the results show that there is a correlation between density and compressive strength values of bio-based mortar samples, linear correlation coefficient R^2 = 0,867 was calculated, which means very strong correlation.

Correlation is fitted by the most suitable function and the equation is reported in the graph including the reliability coefficient R^2.

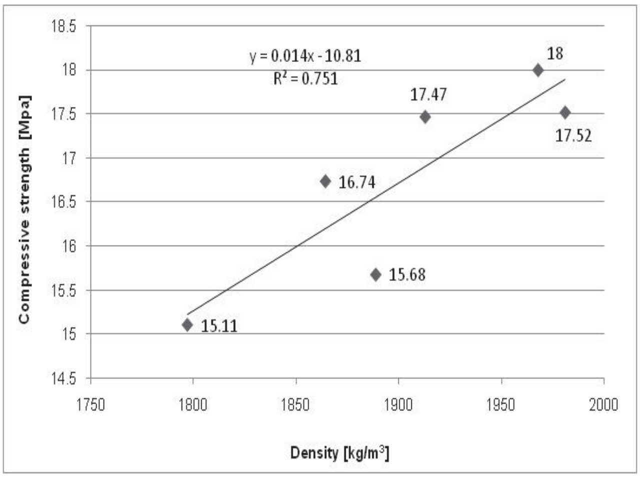


Fig. 7 Dependence of density values on compressive strength of composite samples

The short-term water absorption (1h) values of prepared mortar composite samples, shown in Fig. 8, present increasing water absorbability of composites based on cellulosic fibers and lignocellulosic aggregates with increasing percentage of organic material. This increase is caused by nature of bio-based composites reinforcement. Water absorbability in hardened composites is dependent on internal pore system.

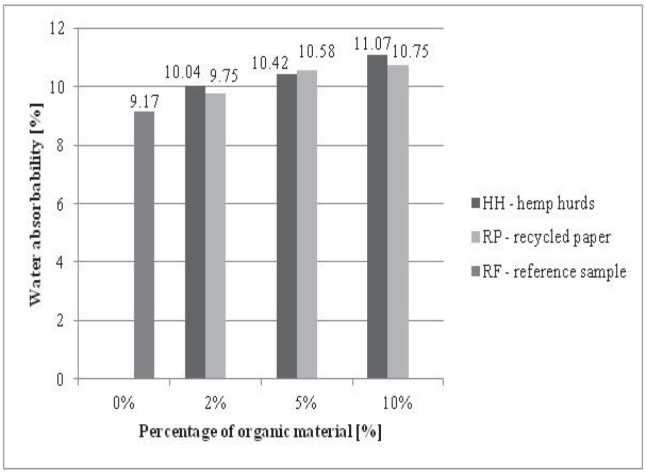


Fig. 8 Influence of organic filler content on water absorbability

IV. Conclusion

This paper deals with use of bio-based material as reinforcement into mortar samples to create sustainable building materials.

The effect of waste cellulosic fibers and lignocellulosic aggregates addition on the physico – mechanical properties of cement based mortars was investigated. Furthermore, the effect of fiber origin (recycled fibers coming from waste paper and hemp hurds as waste of hemp stem processing), its morphologic structure and various portions (2.0%, 5.0% and 10% wt.) of organic material addition to the composite mixture were analyzed.

Cellulosic and lignocellulosic samples coming from various sources showed differences in their morphology (surfaces impurities), structure and chemical composition. Physical and mechanical parameters of bio-based cement mortar composites (density, compressive strength and water absorbability) after 28 days of hardening were tested. The measurements demonstrated that physical and mechanical properties depend on the fiber nature, surface morphology and structure, shape and amount of used organic material into cement based mortar mixtures. The density decrease of bio-based cement-mortar samples is mainly attributed to the fact that used organic composites reinforcement; recycled fibers as well as hemp hurds are light in weight due to their porous structure and nature. However, precisely because of the porous structure of the material leads to increases of water absorbability. The relationship between compressive strength and density of bio-based cement mortar specimens was shown.

Performance is positive in terms of future use of cellulosic fibers and lignocellulosic aggregates into cement-based mortars. The composites on the recycled and renewable material source are potentially contributing to sustainable development due to their environmental benefits.

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