Possibility of cement composite reinforcement by Spanish broom fibres

A considerable attention is currently paid worldwide to the use of cellulose fibres in composite materials. Attempts are made to find even better, environmentally more acceptable, less expensive, renewable and, if possible, locally available replacement for traditional synthetic fibres. The quality of natural fibres is greatly dependent on their origin, processing requirement, composition, and physical properties. Results obtained by testing possibilities of using fibres of the wild plant called Spanish broom, collected locally for cement mortar reinforcement, are presented in the paper. The results show that the quality of Spanish broom fibres is not inferior to the quality of known hemp and flax fibres.

Key words:
Spanish broom, natural fibres, cement mortar, composite strength
1. Introduction

A composite is a combination of at least two different materials that are joined together to form a new, different material, exhibiting greatly improved physical or chemical properties of its individual components, or properties that the individual components do not have. Composite materials reinforced with fibres are characterised by an increased ductility, bending strength, tensile strength, fatigue strength, dynamic strength, and impact resistance. Fibres are short elements exhibiting an increased tensile strength. They are characterised by their aspect ratio, i.e. by the length to diameter ratio. By adding fibres to the matrix of a brittle material such as concrete, the spreading of microcracks is delayed by transfer of stress from the crack position to neighbouring sections[1]. Fibres are classified into steel fibres, synthetic fibres, and natural fibres. Synthetic fibres are divided into organic and inorganic, while natural fibres are classified into vegetable fibres, animal fibres, and mineral fibres. Consumption of non-renewable sources of raw materials, and release of 30% of the total CO₂ emissions, has increased environmental awareness so that, in recent times, and especially in the field of materials, the need has increasingly been emphasized to adopt sustainable development practices and to use environmentally sound materials. The production of synthetic fibres, involving the use of expensive machines and considerable investments, is mostly concentrated in developed countries. On the other hand, natural fibres can be manufactured through human labour using traditional knowledge and practices [2]. Attempts are now increasingly being made to replace steel and synthetic fibres by natural fibres made of jute, sisal, flax, hemp, banana, palm, henequen, kenaf, ramie, cotton, and other types of cellulose fibres [3].

An advantage of natural fibres over other fibres is that they are locally available, economical, biodegradable, renewable, characterised by low density, safer for handling and manufacturing, they are not abrasive and hence cause lesser wear of equipment, they are not conductors, etc., while at the same time, they have all mechanical properties that are needed for improving the composite. Natural fibres are formed through the process of photosynthesis in which plants take carbon dioxide and release oxygen. The use of cellulose fibres results in reduced plastic shrinkage, higher thermal and sound insulation, and in damping vibrations in cement composites. Cellulose fibres also exhibit some disadvantages: the problem of durability, proportioning limitations, and variations in properties. Current knowledge on the durability of fibres points to the need to conduct thorough testing prior to any serious production of cement composites reinforced with natural fibres. Decomposition of fibres in cement composite occurs due to the influence of alkalis, which dissolve lignin and hemicellulose. The influence of alkalis on the disintegration of fibres can be reduced by replacing a part of cement in the composite with fly ash, metakaolin, silica fume, slag, ash obtained by combustion of rice husks, and similar materials. Sometimes even a low quantity of alkalis in cement composite is not sufficient to prevent decomposition of lignin. Fibres can also be treated before being added to the composite, in order to remove the parts of fibres that are sensitive to alkali action. Fibres can be coated with water-repelling substances or they can be impregnated with sodium silicate, sodium sulphite, or magnesium sulphate. Current experience on fibre coating with silane points to an increase in durability of concrete strengthened with natural fibres, and also to an increase in tensile strength and flexural strength. Alkali content of cement composite can be reduced by accelerated carbonation, in the scope of which lime with carbon dioxide forms calcium carbonate CaCO₃. This procedure also influences durability and properties of the composite [4,5].

The fibre content in fibre-reinforced concrete ranges from 0.2 to 2%. Larger fibre content causes difficulties in concrete mixing and placing, and raises the problem of uniform distribution of fibres in concrete. In practice, fibres are added to the mixer containing concrete that has already been mixed. The quality of natural fibres depends on a number of factors: chemical composition, diameter and geometry of fibres, length, surface roughness, fibre processing method, and geographic and weather conditions. Limited usability of these fibres is also due to lower maximum temperature of processing, and hydrophilic properties. Natural fibres exhibit a high tensile strength and low elastic modulus. As to their composition, cellulose fibres mostly consist of cellulose, hemicellulose, lignin, vax, and pectin. Fibre properties greatly depend on chemical composition, especially on the content of cellulose, hemicellulose and lignin. Cellulose is the strongest and stiffer part of fibre that makes it stronger and more durable. Cellulose fibres are polar and hydrophilic, and they absorb a large quantity of water, while their resistance to moisture is low. This is often the cause of a poor contact zone between fibres and the matrix and, consequently, of poor mechanical properties of the composite, especially if the matrix is hydrophobic such as in the case of polymers. Hemicellulose is soluble in water and hygroscopic. It is bound to cellulose by strong hydrogen bonds, and can disintegrate at high temperatures. Hemicellulose forms the bond between cellulose and lignin and, without lignin, plants are not able to grow. A greater quantity of lignin negatively affects tensile strength and elastic modulus [5].

Industry is increasingly interested in the use of environmentally sound low-weight materials of appropriate durability that exhibit satisfactory mechanical properties. For that reason, cellulose fibres are more and more in the focus of interest of industry operators, thanks to their biodegradability, easy processing, and low density. The interest for strengthening materials with natural fibres is expected to grow over the next period. Current worldwide annual production of natural fibres ranges from 70,000 (abaca) to 75,000,000 tons (sugar cane) [6].

The following is written at the FAO pages [2]: Although climate differs from country to country, the variety in natural fibres is broad, resulting in the fact that in every climate zone (with the exception of the extreme dry- and cold climates) at least one fibre crop can grow. This has motivated the authors of this paper to examine to possibility of using Spanish broom, a plant that is widely spread in the Mediterranean area, as a reinforcement in cement composite.

2. Spanish broom (weaver’s broom, Spartium junceum L.)

2.1. Distribution and history

The use of Spanish broom can be traced back to the times of ancient Greeks, Romans, and Carthaginians [7]. It is now believed...
that the city of Split obtained its name from the Greek word Asphalatos, which means Spanish broom field. Spanish broom is a bushy plant that is widely spread in the Mediterranean area and, following the discovery of America, it has further spread to the north and south of America, and it also grows in south-western Asia, and north-western Africa. While it is treated in the USA and New Zealand as a weed, it has always been widely used in the Mediterranean area. It was used as a raw material for textiles, as a vine tying material, for the fabrication of rope, baskets, bags, nets, and it was also used as a roof covering material. It is harvested both as a wild plant (Figure 1) and as a cultivated species. Spanish broom was also used in the fabrication of brooms and hence its English name Spanish broom.

Figure 1. Spanish broom

According to [7] Spanish broom has been planted in Spain, France, and Italy, in places where hemp and flax could not grow due to very poor soil because, as it is written in [8] Thanks to its modest needs Spanish broom, as a wild plant prospering on unfertile rocky terrains of hot and dry coastal areas and islands of the Mediterranean, has outlived flax and hemp. It is not affected by pollution and can withstand temperatures as low as -20°C. The life span of Spanish broom extends to about 25 years.

In Croatia, Spanish broom grows at the Adriatic coast and on islands. As it is a wild plant, its distribution is nonuniform. In addition to Split, some other towns and villages have also been named after this plant: Žanestra (Umag), Brništrovica (Cres), Žukovac (Mljet), Žukova (Hvar), Žukova (Korčula), and Brništrovac (Mljet). In the past, Spanish broom was most often used in the vicinity of Šibenik (Vodice, Pirovac, Zaton) where it was spun and woven. In the 1950s, Spanish broom fibres were processed at as many as 30 locations along the Adriatic area. It is interesting to note that there are areas in this region that are completely devoid of Spanish broom, such as the islands of Lastovo and Vis [8]. A Spanish broom processing plant was opened in the early twentieth century in the town of Omis, but the industrial processing was soon discontinued due to poor management of the processing procedure. Attempts were also made to use Spanish broom for the fabrication of paper in a paper mill in Rijeka. In this respect, preliminary tests for industrial processing of Spanish broom were conducted at Sušak, and plans for future processing units were prepared.

After the second world war, an initiative was started to purchase dry suckers of Spanish broom for the textile and paper industry, and several small factories were built to this effect in Vodice, Zakućan and Opuzen. However, it soon became clear that preparation and processing of Spanish broom is a highly demanding activity, which is why other industrial plants were used instead [8].

Spanish broom loses its significance as a textile raw material in the 1960s. At that time, weaving with Spanish broom was still operated at ten locations, while in Vir, Olib, Ist, Filip Jakov, Biograd, and Betina, this plant was processed to form fibres. The mentioned six fibre processing locations accounted for only 20 % of the total processing activity operated in the mid-1950s.

2.2. Properties of Spanish broom and use of fibres in composites

Spanish broom mostly grows as a bush varying from 1 to 1.5 m in height, although much bigger plants of up to and even in excess of 3 m in height have been found. The roots are strong and well developed, and so Spanish broom is also used for protection against erosion. Similar to other Leguminosae, Spanish broom enriches soil with nitrogen. The plant can grow in clayey, calcareous, neutral, acid, and alkaline soil. It is widely known for its yellow flowers with a very intensive scent (Figure 2) that develop in late spring. By its scent, it highly attracts bees, which is why this plant is very important for apiculture. Flowers have been used for medicinal purposes, and to fill pillows and mattresses, but also in the production of pigments and perfumes.

Figure 2. Spanish broom in full bloom, as observed on the island of Krk (photo by courtesy of Dr Biserka Perković)
Branches are very sturdy and constitute the most important part of the plant, Figure 3. They are composed of two layers: external and internal. While the internal layer is stiff and woody with a porous centre, the external layer is formed of two types of resilient fibrous cells linked with pectinous lamellas into bundles (elementary fibre), and of bundles interconnected with lignin along the branch (technical fibre) [8]. Elementary fibres of Spanish broom are of regular diameter ranging from 5 to 10 micrometers, while the bundle diameter measures approximately 50 micrometers [9, 10].

Spanish broom suckers have a high cellulose content, and the value depends on the way in which fibres are obtained. Spanish broom properties compared to some other widely known fibres are shown in Table 1. According to Table 1, Spanish broom has the lowest specific weight compared to other plants, and the tensile strength value is within the range of values obtained for most natural fibres. The study of Spanish broom fibre quality is mainly oriented toward this plant’s use in textile industry, but there are studies in which Spanish broom is regarded as reinforcement of composites and, in this respect, a special focus is currently being placed on biopolymer study. Thus some research has been published on the use of Spanish broom fibres as a reinforcement of composite materials.

| Plant type   | Specific weight | Tensile strength [MPa] |
|--------------|-----------------|------------------------|
| Spanish broom| 0.95–1.05       | 400–750                |
| Flax         | 1.40–1.52       | 500–900                |
| Hemp         | 1.42–1.54       | 310–750                |
| Cotton       | 1.5–1.6         | 300–700                |
| Jute         | 1.44–1.53       | 320–800                |
| Kenaf        | 1.4             | 223–930                |
| Ramie        | 1.0–1.55        | 400–1000               |
| Sisal        | 1.33–1.5        | 363–700                |

Angelini et al. [9, 10] compared Spanish broom fibres with ramie fibres, and artificial fibres. Spanish broom fibres exhibit somewhat poorer mechanical properties compared to ramie, as shown in Figure 4.a.

Figure 3. Spanish broom shoot (left) and longitudinal section of Spanish broom plant viewed by microscope Opton Axioskop MC 63A: 2 - sclerenchyma, 3 – parenchyma with chloroplasts (right)

Figure 4. a) stress-strain diagram; b) influence of gauge length on the strength of Spanish broom, ramie, carbon and glass fibres [9]
The stress–strain diagram of both fibres is almost linear up to the failure point. The elastic modulus for ramie amounts to approximately 65, while it is approximately 21.5 GPa for Spanish broom, which is within the range of the fibres of cotton (5.5 - 12.6 GPa), jute (30 GPa), sisal (9 - 38 GPa [3]), and similar cellulose fibres. The authors emphasize that, although the elastic modulus of Spanish broom accounts for only 30 % of the corresponding value for ramie, it is still higher compared to some non-oriented polymers where this value varies between 1 and 3 GPa. Thus Spanish broom is considered favourable for reinforcing some composite materials. According to Figure 4 (right), the comparison of these two cellulose fibres with artificial fibres of carbon and E-glass shows that artificial fibres are stronger than cellulose ones, but that their specific weight is higher, which points to the conclusion that both cellulose fibres can be used as reinforcement, at least in non-structural applications. The figure shows that all four types of fibres exhibit the same trend, i.e. an increase in test fibre length results in a decrease of strength. In addition, the authors have shown that the strength at the contact between cellulose fibres and epoxy resin matrix is by 1.4 to 2.2 higher compared to artificial fibres [9].

Nekkaa et al. [11] investigated water absorption of the composite made of polypropylene matrix and Spanish broom fibres. Once the samples had been dried to the constant mass at 70 °C, they were placed in the distilled water at the temperatures of 23 and 85 °C. The objective of the testing was to determine the influence of the proportion of Spanish broom fibres, influence of treatment of fibre surface with silane, and the influence of temperature on the quantity of water absorbed by the composite. It was established that the quantity of absorbed water increased with an increase in Spanish broom proportion in the composite. The composite containing treated Spanish broom fibres is characterized by lower absorption compared to the composite with untreated fibres. The impact strength of the composite greatly depends on the quantity of absorbed water, and water saturated samples exhibit a low impact strength. In case of fibres treated with silane, images obtained by scanning microscope show reduced surface roughness of fibres, which enables good adhesion between the fibres and the polypropylene matrix.

Picuno [12] tried to strengthen adobe bricks with Spanish broom fibres. The adobe brick is a traditional type of brick that is dried in the sun, and is made of clay. It is usually formed as a mixture of coarse sand, raw clay soil, and lime. The addition of fibres lowers the incidence of brick cracking while it is being dried in the sun. The author formed 15-cm cube-shaped samples containing 49.3 % of clay, 36.9 % of sediments (silt), and 13.8 % of sand. Natural fibres accounting for 1/3 of the volume were added to the mixture, and four groups of samples were formed according to the type and fibre adding method: samples reinforced with wheat straws: randomly disposed across the volume or disposed orthogonally to the compression load, and samples reinforced with Spanish broom: which is randomly disposed across the volume or disposed orthogonally to the compression load. The compressive strength values roughly correspond to the strength of the adobe brick that has not been reinforced. The author tested tensile strength of Spanish broom, as a natural sprig and as a spun rope. The tensile strength of the Spanish natural sprig amounts to $41.53 \pm 4.13$ N/mm², while the corresponding strength of the rope amounts to $36.32 \pm 6.37$ N/mm². In conclusion, the author points out that, in the category of natural fibres, Spanish broom has a potential for improving the adobe brick, although further research is needed to determine in what way this potential can be realized.

Spanish broom fibres are still the subject of study in Italy. As the greatest objection to the use of Spanish broom is the way in which fibres are obtained, the authors [13] from Calabrian High Tech S.r.l patented a fibre manufacturing device. There are six phases through which Spanish broom bush must pass to obtain the desired quality of fibres. Figure 5 shows bush position in the first phase of processing. The idea is that one worker starts the manufacturing process by hanging a Spanish broom bush in the frame, and the ensuing procedure is fully automated. In the initial phase, four bushes are processed in parallel, but the number of places at the support is soon to be doubled. Although the plant has already started operating, the quantity of fibres produced is still insufficient, and so attempts are currently being made to improve the manufacturing process.

3. Experimental part

Cement mortar prisms reinforced with Spanish broom fibres were prepared in the experimental part. The cement CEM I 42.5 R and CEN standard sand according to EN 196-1 were used. A bag with standard sand (1350 g), cement (450 g) and water (225 g) is needed for the preparation of a series of three prisms. The mortar is mixed in an automatic mortar mixer. After the cement and
water have been mixed for 30 seconds, the sand is automatically added during the ensuing 30 seconds of mixing, which is followed by 30 second mortar mixing at a greater operating speed. After that the standard mixing program is stopped, the vessel is taken out of the mixer, Spanish broom fibres are added, and process continues with manual mixing. Spanish broom fibres are shown in their original length in Figure 6.

Figure 6. Spanish broom fibres

They were cut into three lengths for the testing: 10 ± 2 mm; 20 ± 2 mm, and 30 ± 2 mm. Depending on fibre length, the samples were marked as B1, B2 and B3 where the number denotes fibre length in cm. Fibres were added to the cement mortar in the quantity of 0.5 and 1 % of the total volume. Samples with 0.5 % of fibres were marked with -05 and those with 1 % of fibres were marked with -1. Two series of samples with fibres 10 mm in length were made in the quantity of 0.25 and 0.75 % of the volume.

The workability of fresh mortar was tested using the flow method. The mould shaped as truncated cone was filled with mortar in two layers. Each layer was compacted 10 times, the mould was raised after 15 seconds, and the flow table was raised and lowered 15 time at the rate of one time per second. An average mean value of mortar diameter was determined in two perpendicular directions, and the results obtained were presented and evaluated in Table 2.

Table 2. Workability of mortar

| Sample  | Consistence (mm) | Category |
|---------|------------------|----------|
| Etalon  | 188              | plastic  |
| B1-05   | 135              | stiff    |
| B1-1    | 140 (loose)      | stiff    |
| B2-05   | 138              | stiff    |
| B2-1    | 125              | stiff    |
| B3-05   | 153              | plastic  |
| B3-1    | 130              | stiff    |
| B1-025  | 141              | plastic  |
| B1-075  | 133 (loose)      | stiff    |

Figure 7 shows the drop in workability of mortars B1-05 and B1-1 as related to etalon (standard), which could have been expected as the quantity of water in samples was kept constant. The results for mixture B1-1 are numerically better compared to mixture B1-05, but it can be seen in the figure that its workability is poorer, and that it relates to dry consistence. According to Table 2, fibre length does not affect workability, but rather workability is affected by fibre content. A similar trend of drop in workability was obtained by Islam et al. in paper [14], where comparison is made between ordinary concrete and high-strength concrete reinforced with steel and natural coconut fibres. The fibre quantity amounted to

Figure 7. Workability of mortar without fibres, with 0.5 % of fibres, and with 1 % of fibres
Introduction

0.5 and 1 % of the volume, just like in this paper. By slump test conducted for both types of concrete, they established that the drop in workability is greater for the higher fibre content, and that the drop in workability in case of reinforcement with natural fibres is much greater compared to steel fibres. For instance, for both concrete types, the slump was reduced by 92 % for 1 % of coconut fibre content. Mortar was then placed into three gang moulds on the vibrating table. After being kept 24 hours in a wet chamber, specimens were demoulded and placed in water, where they were cured for 27 days following the day of the testing. Flexural strength test results are shown in figures 8 and 9.

Compressive strength results, shown in figures 10 and 11, reveal that the results are influenced by fibre content but also by fibre length. The strength decreases with an increase in fibre content. As it is more difficult to compact and place mortar with fibres, the resulting concrete exhibits greater porosity, and so the result obtained could have been expected.

According to Figure 8, samples with Spanish broom fibres exhibit a somewhat lower flexural strength compared to the etalon. The lowest values were obtained for samples B1-1, B2-05 and B2-1, whose flexural strength values are lower by 10.6 to 12.8 % compared to the etalon, while the values closest to the etalon were obtained for samples B3-05, B1-025, and B1-075. Figure 9 shows the influence of length and quantity of fibres on flexural strength.

According to diagram, a higher content of fibres results in lower strength values, while the fibres 10 and 30 mm in length exhibit better results compared to fibres 20 mm in length. Kiran et al. obtained in [15] an optimum length of natural fibres of about 30 mm. By testing tensile strength of the composite reinforced with the fibres of hemp, banana and sisal, they established that the strength increases with an increase in fibre length from 10 mm to approximately 32 mm, while the strength decreases with further increase in fibre length until 70 mm.

The relationship between the strength and fibre content is shown for samples with fibres 10 mm in length. Both strength values continuously decrease with an increase in fibre content, but a deviation was noted at sample B1-075 with regard to flexural strength.

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the scope of which authors tested cement mortar prepared with hemp and flax fibres. Merta et al. [16] prepared cement mortar of a somewhat different composition, and the compressive and flexural strength results are presented in a relative relationship to the etalon. Compressive strength results are shown in Figure 13, and flexural strength results are given in Figure 14. According to Figure 13, better compressive strength results were obtained for Spanish broom samples proportioned at 0.5% of the total volume, when compared to all fibres proportioned at 1% of the volume. Sample B1-1 is closer to the result obtained for hemp, while the lowest strength is exhibited by flax. The flexural strength for sample B1-05 is closer to the results obtained for hemp and flax, Figure 14.

Dry samples were also tested in the scope of the research published by Nekkaa et al. [11] and in [8], where the difference between the dry and wet Spanish broom samples was considered. After 28 days, the samples were taken out of the water and were then kept for seven days in a room at the temperature of 20°C and at relative humidity of 50%. The results obtained are presented in figures 15 and 16. According to Figure 15, the same principle can be observed for both dry and wet samples subjected to compressive strength testing. In the case of flexural strength testing (Figure 16), dry sample B1-1 exhibits better results in dry compared to wet state, while sample B1-075 with the highest wet strength, exhibits the greatest fall in dry state. As samples for dry and wet testing were prepared in the same recipient, it is possible that fibres were unevenly distributed for the two sample series. The sample B3-05 exhibits good results in both dry and wet state, which is in accordance with [15]. Although short fibres do not generally contribute to an increase in sample strength, their contribution should consist in a higher ductility of the material. This testing for samples with Spanish broom fibres still remains to be conducted by the authors of this paper.

4. Conclusion

Spanish broom is a wild bushy plant typical for the Mediterranean area, where it used to have a much greater significance in former times. In fact, it was used as a raw material for textile, for fabrication of footwear, ropes, sails, nets, baskets, etc. With the advent of industrial plants such as hemp and flax, the intensity of its use gradually decreased. Compared to flax and hemp, Spanish broom is much easier to cultivate as it is not demanding as to selection of soil and, more importantly, once
Spanish broom fibres can also be used in composite materials, and it is in this paper that the possibility of cement composite reinforcement with Spanish broom fibres has been tested for the first time.

According to the results obtained in the paper, and comparisons with other cellulose fibres, it was established that Spanish broom fibres can potentially be used as a reinforcement agent in cement composites. It was also revealed that fibres 10 and 30 mm in length have the highest potential for increasing compressive strength. A more detailed investigation should be conducted in order to find an optimum combination of the quantity and length of Spanish broom fibres in cement matrix. It is currently planned to test influence of fibres on the ductility of concrete/mortar, which would actually be the primary role of short fibres in such composites.

It is perhaps the price of Spanish broom fibre manufacturing that has reduced the competitive edge of this plant compared to other types of natural fibres. However, thanks to current encouragement of sustainable construction practices, and under appropriate conditions, the manufacture of these fibres could become of interest in the near future. This idea involves use of cultivated Spanish broom, as the original wild plant is a protected species. The role of this plant in composites can be defined in relation to the improvement of environment, protection of roadways and rail routes, forestation and other possible applications, especially following further development of the fibre separation process, which is now being studied quite intensively. It should finally be noted that Spanish broom is a plant deserving greater attention.

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