sustainability

MDPI

Article

Preliminary Research on Moss-Based Biocomposites as an Alternative Substrate in Moss Walls

Rafael Alvarez Gutiérrez [1,*], Johan Blom [1], Bert Belmans [1,2], Anouk De Bock [1], Lars Van den Bergh [1] and Amaryllis Audenaert [1]

[1] EMIB Research Group, Faculty of Applied Engineering, University of Antwerp, Groenenborgerlaan 171, B-2020 Antwerp, Belgium; johan.blom@uantwerpen.be (J.B.); bert.belmans@uantwerpen.be (B.B.); anouk.debock@uantwerpen.be (A.D.B.); amaryllis.audenaert@uantwerpen.be (A.A.)

[2] Architectural Engineering Research Group, Department of Architectural Engineering, Vrije Universiteit Brussel, Pleinlaan 2, B-1050 Brussel, Belgium

* Correspondence: rafael.alvarezgutierrez@uantwerpen.be

Abstract: Addressing urban air pollution is a pressing challenge, prompting the exploration of mitigation strategies such as urban greening. However, certain innovative greening approaches, while promising, may inadvertently incorporate unsustainable elements that undermine their eco-friendly philosophy. In this context, our research focuses on addressing the replacement of a petroleum-based filter substrate in an existing 'green' outdoor air purification system that utilizes 'moss filters', known as a 'moss wall'. This initiative is driven by concerns about microplastic leakage from the substrate and the need to optimize the moss wall system in terms of circularity. This preliminary study presents a crucial first step, aiming to assess the feasibility of developing a circular, bio-based plate as a replacement for the existing microfiber filter substrate. The focus is on the potential of this plate to recycle moss from the system itself as raw material, ensuring structural integrity and the ability to support its own weight. To achieve this goal, a series of controlled experiments were conducted in a laboratory setting using cellulose, corn starch, and metakaolin binders. Our findings indicated that cellulose was crucial for the structural integrity, starch significantly enhanced the sample strength, and metakaolin improved the water resistance. These insights culminated in the creation of a laboratory-scale moss-based composite prototype, with moss constituting more than half of the total mass. This prototype demonstrated promising results as a starting point for a more environmentally friendly and bio-based moss wall substrate. Subsequent research efforts will concentrate on optimizing the binder and fiber composition, evaluating and improving the bioreceptivity and filter properties, conducting outdoor testing, and scaling up the prototype for practical implementation.

Keywords: biocomposites; sphagnum moss; moss wall; natural fibers; bio-binders; air purification; bio-based materials

Citation: Alvarez Gutiérrez, R.; Blom, J.; Belmans, B.; De Bock, A.; Van den Bergh, L.; Audenaert, A. Preliminary Research on Moss-Based Biocomposites as an Alternative Substrate in Moss Walls. *Sustainability* **2023**, *15*, 16500. https://doi.org/10.3390/su152316500

Academic Editor: Giouli Mihalakakou

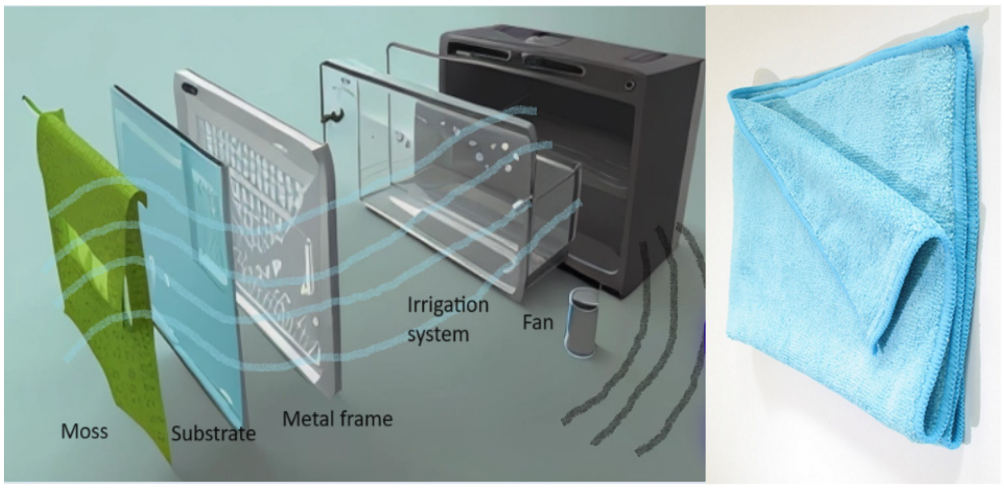
Received: 26 October 2023
Revised: 22 November 2023
Accepted: 30 November 2023
Published: 2 December 2023

1. Introduction

Being exposed to ambient air pollution is a severe threat to human health causing around 4.2 million deaths every year [1–4]. Particulate matter (PM) is the main air pollutant in urban environments that causes severe health problems [2,3,5]. These small particles, solids or liquid aerosols, come from a variety of sources, both natural and anthropogenic, such as fossil fuel combustion, construction dust, and industrial processes [6,7]. Mainly, two types are defined based on particle diameter: PM10 are particles with a diameter of 10 µm or less and PM2.5 are particles with a diameter of 2.5 µm. These particles, especially PM2.5, can penetrate deep into the lungs, leading to severe health issues [3,5].

One way to combat air pollution is to address the problem at its source and reduce emissions from polluting activities. Another way is to filter polluted air using active or passive filtration technology [8–12]. Various methods and systems have been introduced to actively filter polluted outdoor air [8,11,12]. Among these are solutions that make use of the

air-purifying properties of plants. One such method is the use of 'moss walls' [11,12]. These systems, illustrated in Figure 1a, draw in polluted air through ventilation fans installed inside a hollow wall that is covered with 'moss filters', passing the stale air through the moss and the substrate on which the moss grows. After filtering, the purified air is released back into the environment. Due to the absence of a developed root system mosses have developed a very high surface area of densely packed leaves to absorb water and nutrients compared to other plants [13,14]. This allows them to trap air pollutants, such as PM, in a very efficient way, similar to manmade filters [15]. Research performed in 2019 at the University of Antwerp has shown that the filtration efficiency of moss panels is 43.1% for PM10 and 22.8% for PM2.5 [16].



(**a**) (**b**)

Figure 1. (**a**) Exploded view of existing moss wall; (**b**) current substrate: synthetic microfiber cloth. The substrate is light blue in color in both images.

To use living moss as part of a filter setup in an air purification system, it needs a breathable and moisture-retaining substrate on which it can grow. Currently, in the moss wall that is the subject of this study, a synthetic microfiber cloth is used for this purpose, see Figure 1b [11,16]. However, this material is highly environmentally polluting as it contains and releases microplastics to the environment, i.e., pieces of plastic less than 5 mm in diameter, that are not degradable and are harmful to various ecosystems [17]. The exposure of microplastics to humans, whether through ingestion or inhalation, can cause potential health risks. These risks include medical conditions, such as gastrointestinal issues, chronic pulmonary diseases, fibrosis in the lungs, and an elevated risk of lung cancer [18,19].

To mitigate the entry of microplastics from the substrate of the moss wall into the environment, this study aims to lay the foundations for a bio-based alternative to microfiber cloth, utilizing excess moss from the system as a raw material. By using saturated moss from the filters and/or excess moss (after cleaning), the system will become more self-sustaining with reduced dependence on external inputs. Therefore, this solution not only addresses immediate environmental concerns but also promotes the circularity of the moss wall. This preliminary study primarily focusses on assessing the feasibility of developing such a biocomposite, placing emphasis on structural integrity and the material's ability to support its own weight, while thoroughly exploring associated materials and methods. At this stage, the scope of this research does not extend to evaluating and optimizing the applicability of the biocomposite as a substrate or its filtration properties.

2. State-of-the-Art Biocomposite Materials

Biocomposites are materials consisting of a reinforcing phase and a matrix surrounding the reinforcing phase, where at least one of the two is derived from natural sources. They can be categorized based on the type of matrix and fibers they employ, resulting in two

main classifications: partially biodegradable and completely biodegradable. Within the category of completely biodegradable biocomposites, bio-fibers are utilized in combination with matrices composed of biodegradable polymers. However, currently, the market for fiber reinforcement is still dominated by synthetic fibers, resulting in mainly partially biodegradable solutions [20]. In many applications, natural fibers are a good alternative to synthetic fibers that are derived from petroleum or other non-renewable resources. This is supported by extended research on the mechanical properties of natural fibers, which exhibit promising similarities to traditional fibers but still have room for further improvement [21]. They are considered to be more environmentally friendly as they are made from renewable resources. Moreover, they have low CO_2 emissions during processing, are abundantly available and cheap, and can be recycled or biodegraded at the end of their life [22]. By utilizing these materials, biocomposites can offer a sustainable solution to the disposal management issues faced by certain countries, addressing waste accumulation while promoting resource efficiency [23].

Much progress has already been made in developing methods to make biocomposites using different combinations of natural fibers and binders. In this respect, several recent studies have explored the use of moss-based biocomposites for constructing thermal insulation plates in buildings [24–26]. In these studies, *Sphagnum* moss was used as the main component in combination with various binders, viz., animal glue, liquid glass, and urea-formaldehyde. Biocomposites for thermal insulation applications have also been produced from hemp and flax shives with potato starch as a binder. The aggregate–binder ratio was chosen to be the lowest possible so the sample could withstand its self-weight, resulting in a ratio of 1:3. Starch was prepared using the casting method, whereafter the mixture was compressed and left to dry [21]. Starch is also commonly used in the packaging industry for the preparation of bioplastics. As shown by Marichelvam et al. (2019), the tensile properties of starch are suitable for the production of packing materials, and the preparation method is straightforward [27]. Furthermore, several papers have investigated the use of metakaolin in combination with corn starch to create a biofilm. This combination results in an improvement in the mechanical tensile strength and hydric properties of the obtained biocomposites [28,29]. The use of wood and paper pulp as binders in biocomposites is currently also being researched [30]. Fuentes et al. (2021) developed biodegradable pots based on different waste materials such as, among others, used paper, wheat flour, and corn-waste flour [31]. Additionally, a combination of wooden fibers and peat moss has been used for the design of biodegradable pots, containing 80% wooden fibers and 20% peat moss, to improve the uptake of water [32].

To conclude, the growing interest in biocomposites offers numerous opportunities to develop sustainable materials for diverse applications. Although there is a lot of ongoing research on natural fibers and biodegradable polymers to replace petroleum-based products, the development of fully biodegradable biocomposites is still in its early stages. Moreover, the integration of moss fibers alongside other natural fibers and biodegradable polymers to develop biocomposites, other than for insulation purposes, remains a relatively unexplored topic. In the broader context of continuous research on moss walls for urban air purification, and with the aim of advancing the state of the art in (moss-based) biocomposites, this study therefore explores the feasibility of creating a new fully biodegradable, moss-based biocomposite. In the first instance, the objective is to develop a material that uses excess moss from the system as a raw material and that can support its own weight while preserving its structural integrity. Consequently, this preliminary investigation stands as a meaningful contribution to ongoing research efforts and advancing the current state of the art.

3. Materials and Methods

The previous section highlighted the wide variety of materials available for biocomposites that are currently in active use and exploration. For our specific application as a substrate for moss filters, we adopted a meticulous material selection process, with a strong emphasis on sustainability. Our selection criteria revolved around the following

principles: the chosen raw materials needed to be natural, biodegradable, reusable, renewable, and abundantly available and had to provide a nutrient-rich environment to stimulate moss growth. We also considered the origins of these materials to minimize their environmental footprint, favoring locally sourced options. Furthermore, it was imperative that the materials did not leave any harmful residues that could detrimentally impact the environment.

In addition to material selection, we developed a detailed plan for binder preparation and sample production. With the aim of developing a biocomposite that could support its own weight while preserving structural integrity, various binder-to-fiber ratios were systematically tested to achieve samples with an optimal moss content and structural integrity. Subsequently, we conducted manual assessments and a series of preliminary laboratory-scale tests to provide an initial characterization of the most successful samples.

3.1. Materials

3.1.1. Natural Fibers

The natural fiber used in this study was the moss fiber of *Sphagnum* moss. This moss species is widely available and has already been used in research on moss-based biocomposites for thermal insulation purposes [25,26,33]. Furthermore, this moss species was used as a part of the filter in the moss wall, enabling air purification. With circularity in mind, it would be ideal to use the saturated moss from the filters and/or existing excess moss, after cleaning, as a resource for the new substrate. In this manner, a self-sustaining system can be achieved with minimal external materials. Hence, fibers of *Sphagnum* moss were used as the primary material of the new biocomposite. Being aware of moss fibers being characterized by their relatively short length and limited tensile strength, we acknowledged the necessity for enhanced structural strength. To address this need, we explored the inclusion of longer fibers, such as jute, flax, sisal, and hemp. These longer fibers possess a significantly higher tensile strength when compared to moss fibers. Based on practical considerations, we ultimately opted for jute to fortify the structural integrity of the plate. Despite jute originating primarily from eastern India and Bangladesh [34] and the study being conducted in Antwerp, Belgium, jute was chosen for its accessibility and easy handling.

Preceding the production process, the moss fibers underwent a cleansing process, in which the moss was washed with running water to remove impurities such as sand. Next, the moss fibers were blended in a mixer. The jute fibers were extracted from a jute rope by cutting them into lengths of about 20 to 30 mm and by splitting the rope into four fibers. In two samples, long moss fibers were used instead of jute fibers.

3.1.2. Binders

Three binders were evaluated to increase the strength of the plate. An important requirement was that the binders should not be of animal or synthetic origin. Thereby, animal-based binders such as chitosan, animal glue, and gelatin, frequently used in biocomposites [25,31,35,36], were excluded from the experiments. Cellulose, the first binder, is the main structural component of the cell wall in plants. In this study, the cellulose was derived from biodegradable plant pots composed of paper and wood pulp. These biodegradable pots had a high cellulose content and will be referred to as 'cellulose' in the remainder of the paper for the sake of conciseness.

As a second binder, corn starch was used. This is a carbohydrate composed of semi-crystalline granules, which consist of glucose chains, formed by plants during photosynthesis. The structure of corn starch is initially crystalline as the glucose chains in the granules are connected with hydrogen bonds. Adding water and increasing the temperature causes the granules to absorb water until the bonds between the glucose chains break. The glucose chains bind with the surrounding water molecules, and the density of the mixture increases, achieving the gelatinization of starch. Retrogradation occurs when the mixture

cools down. The glucose chains repel the water molecules and rebind with other glucose chains, regaining a crystalline structure with high mechanical properties.

Metakaolin, as a third binder, is the calcinated form of the clay mineral kaolin, which is a type of soft, white clay found all over the world. The calcination process drives all the water out of the kaolin, which results in metakaolin, an amorphous aluminum-silicate ($Al_2O_3 \cdot 2SiO_2$) [37]. Metakaolin is often used as a partial replacement for cement, which enhances the strength as well as reduces the amount of cement needed in concrete [38].

The cellulose was weighed and included in the mixture, while the corn starch and metakaolin needed additional preparation. The corn starch was synthesized using the casting technique, a widely employed method for generating gelatinized starch, as evidenced in prior works [39–42]. Initially, 15 g (12.5% proportion in the mixture) of corn starch in a granular state was introduced into 150 mL of water and manually stirred until complete dissolution. The mixture was then heated to 80 °C and continuously stirred for 10 min to achieve starch gelatinization (Figure 2a). In the case of metakaolin preparation, the weighed clay in granulated form was initially saturated with water via a gradual addition of water while stirring the mixture until a paste-like consistency was attained (Figure 2b). Weights were measured in a dry state with a measurement accuracy of ±5 g and volumes with a measurement accuracy of ±5 mL.

(**a**) (**b**)

Figure 2. (**a**) Preparation of corn starch; (**b**) preparation of metakaolin.

3.2. Methodology

3.2.1. Sample Composition and Identification

Table 1 gives an overview of the composition of the tested samples. Various combinations of binders, fibers, and lengths of fibers were tested to achieve a strong sample that contained a high moss content. By using these combinations, the samples could be compared, enabling a separate analysis of the effect of each material.

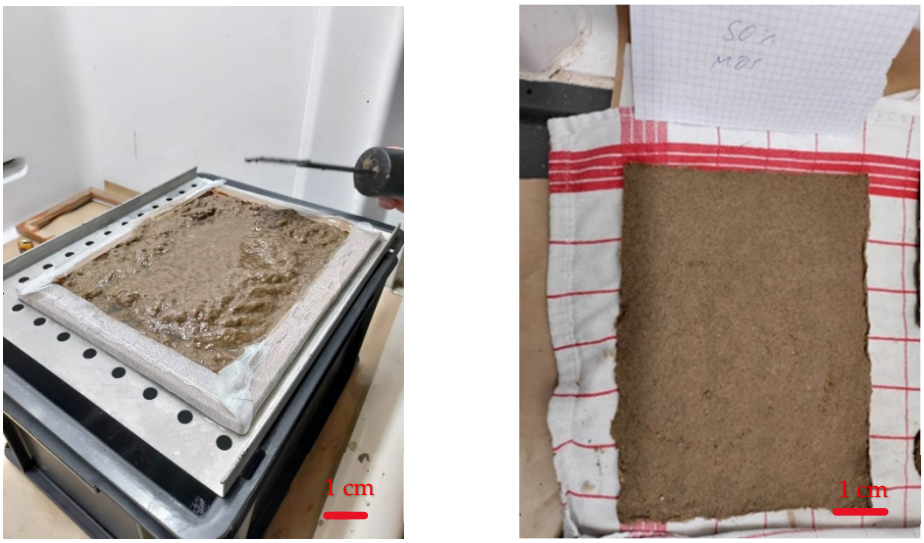
The samples were named based on their composition. The first part of the name 'X/X' refers to the moss/cellulose ratio of the sample. This ratio is followed by a letter when an additive was implemented in the mixture. The letters C, M, LM, and LJ indicate the addition of corn starch, metakaolin, long moss fibers, and long jute fibers, respectively. The letter is followed by a number that corresponds to the weight in grams (g) of the material.

Table 1. Samples tested with different compositions of moss fibers and various binders.

Sample	Moss (g)	Cellulose (g)	Ratio Moss/Cellulose	Corn Starch (g)	Metakaolin (g)	Long Fibers Moss (g)	Long Fibers Jute (g)
50/50	60	60	50/50	-	-	-	-
75/25	90	30	75/25	-	-	-	-
90/10	108	12	90/10	-	-	-	-
100/0	120	-	100/0	-	-	-	-
100/0-C15	120	-	100/0	15	-	-	-
90/10-C15	108	12	90/10	15	-	-	-
100/0-M30	120	-	100/0	-	30	-	-
90/10-M30	108	12	90/10	-	30	-	-
90/10-M60	108	12	90/10	-	60	-	-
90/10-C15-M60	108	12	90/10	15	60	-	-
90/10-LM30	108	12	90/10	-	-	30	-
90/10-LM15-C15	108	12	90/10	15	-	15	-
90/10-LJ5	108	12	90/10	-	-	-	5

3.2.2. Production of Samples

While in many studies the production of biocomposites involves hot pressing and extrusion at high temperatures, this study opted for a more climate-friendly and energy-efficient technique that avoids high temperatures and high energy inputs [43,44]. The production process used was inspired by the production process of paper and nori sheets. In these processes, the wood and seaweed, respectively, are shredded and mixed with water. The mixture is then pressed to remove the excess water, after which the sheet is dried. To form the samples, the moss and binders were mixed in a blender with the addition of water to improve the mixing process. The materials were mixed until a homogeneous mixture was formed. For the shape of the plate, a picture frame of one A4 size was used (Figure 3a). The mixture was placed on the filter cloth, after which as much water as possible was pressed out. Lastly, the plate was turned over and laid to dry on a towel (Figure 3b). After one week, the sample was fully dried out and ready for testing.



(**a**) (**b**)

Figure 3. (**a**) Mixture placed on the filter cloth, ready to press out the excess of water; (**b**) sample laid to dry on a towel.

3.2.3. Evaluation Criteria and Characterization of the Samples

The samples were visually assessed to qualitatively characterize their composition and the adhesion between the binders and fillers, providing a comprehensive overview

of the microstructure. A laser scanning confocal microscope 'Keyence VKX100' (https://www.keyence.com/landing/microscope/pr_vk-x_feauture.jsp, accessed on 6 March 2023) was used for this purpose. The microstructure was studied with a magnification of 90×, which stands for the combination of a magnification objective 18× and a lens magnification of 5×.

The tensile strength was measured with an 'Instron 3366' device and the accompanying software 'Instron Bluehill vers. 2' (https://www.instron.com/-/media/literature-library/products/2011/06/3300-series-table-model.pdf, accessed on 3 April 2023). The starting conditions included no preforce, and the tensile testing speed (extension rate) was set at 5 mm/min. Prior to conducting the tensile test, the samples underwent a drying period of at least one week following production, ensuring that they were thoroughly dried out by the time of testing. This was verified by weighing the samples until a stable weight was reached, as well as measuring the moisture content using a 'PeakTech 5200' moisture meter. Subsequently, three strips measuring 20 ± 1 mm in width and depth varying from 2 ± 1 to 6 ± 1 mm were cut from each sample to undergo testing. The average of the three measurements was taken to obtain a representative value for the tensile strength of the sample composition.

To assess the water retention capacity of the samples, the water permeability was measured. The sample was placed atop a container, ensuring that water could pass through it unhindered by any surface. Subsequently, a tube was affixed onto the sample using silicone and filled with 25 mL of water. The time needed for the water to pass through the sample gave rise to the water permeability. The test was only performed once for each sample at one spot, making the results unreliable. It is therefore important to note this was the first indicative test to provide an idea of the porosity/water permeability of the samples.

To assess the bioreceptivity of moss on the samples, three different techniques were tested. In the first technique, moss was inserted at various points on the sample and loose moss was placed on the surface. Secondly, a mixture composed of 30 mL water, 1 tablespoon of yoghurt, and 10 g of shredded moss fibers was evenly spread on the surfaces [9,45]. The plates were held in boxes covered with plastic film, with small cut-outs for ventilation, to create a humid environment conducive to moss growth. Water was regularly sprayed on the plates to maintain a humid environment, and shading of the moss was ensured. A third option was explored to grow moss on the plates that involved the incorporation of cut-outs and folding the plate forward before drying. Next, soil and untreated moss were inserted in these designated points. Finally, once the sample was fully dried out, it was positioned vertically in a shaded location and sprayed with water every day to monitor the moss growth.

A mold was created with a 3D printer, 'Flashforge Guider IIs' [46], provided by the University of Antwerp, and the accompanying software 'Flashprint vers. 5'. The mold design was taken from Aidan Leitch's creation [47]. The mold was a 6 cm by 6 cm square with a flat base and a wave pattern on top. It consisted of three parts, the base, the walls, and the top. Another dimension explored was the potential to create elements with a form other than the flat A4 size. While synthetic fabric's stiffness resulted from the tensioning forces when suspended in a metal frame, the plate could not rely on the tensile stiffness due to its brittleness. However, the form of the samples can be optimized to enhance the stiffness.

4. Results

4.1. Manual and Visual Assessment of Samples

Manual assessment of the samples provided valuable insights into the different combinations between fibers and binders, as well as the resulting cohesion and mechanical characteristics of the samples. A difference in robustness and stiffness was observed between the four samples solely composed of moss and/or cellulose pulp. The sample '100/0' (Figure 4a), composed entirely of moss, exhibited minimal cohesion among the moss fibers,

as they broke quickly when subjected to a little force. The samples '90/10', '75/25', and '50/50' (Figure A1a) revealed that increasing the cellulose content in the mixture strengthened the plates and increased the stiffness. However, it was observed that the sample '90/10' (Figure 4b), containing a minimal amount of cellulose of 10%, was robust enough to bear its self-weight and maintained its structural integrity.

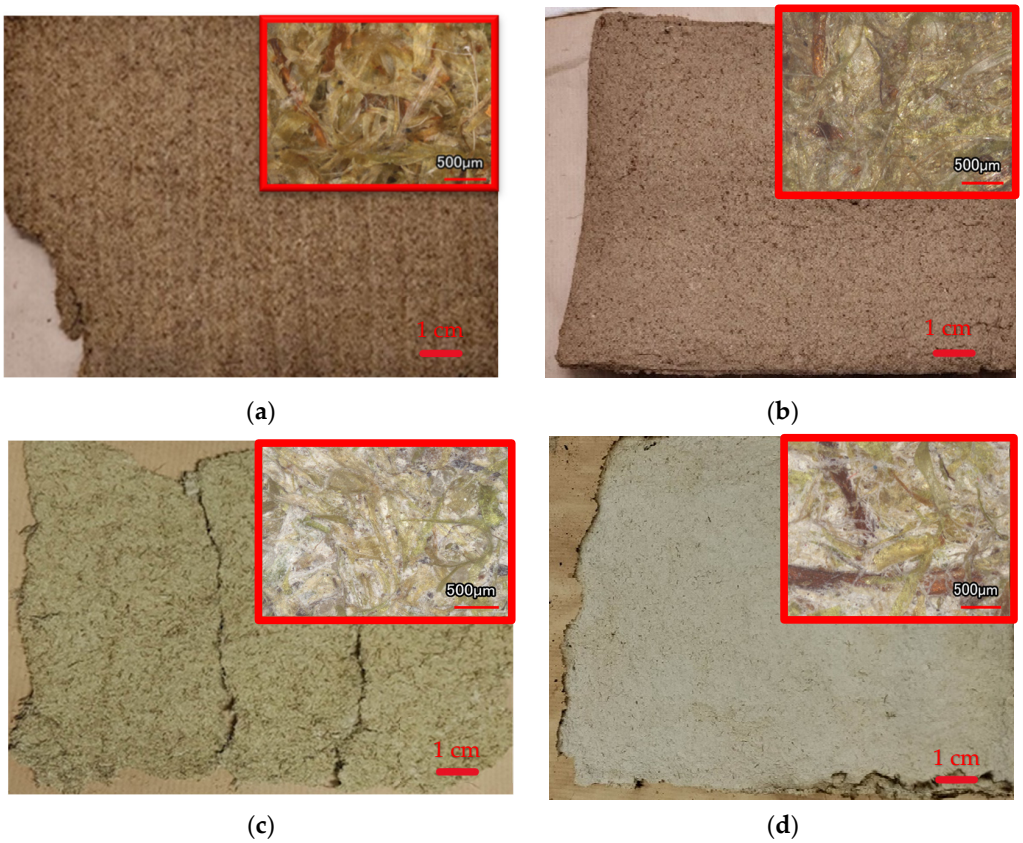


Figure 4. Overview of the samples with different combinations and proportions between fibers and binders, and microscopic image with a magnification of 90×. The composition of the samples is listed in Table 1: (**a**) sample '100/0'; (**b**) sample '90/10'; (**c**) sample '100/0-M60'; (**d**) sample '90/10-C15-M30'.

Next, other binders, such as corn starch and metakaolin, were incorporated into the mixture of the samples. The sample '100/0-C15' (Figure A1b) showed a greater cohesion between the moss fibers compared to sample '100/0'; however, the sample also collapsed when force was applied to it. In contrast, the sample '90/10-C15' (Figure A1c), consisting of moss fibers, cellulose pulp, and corn starch, displayed increased strength and stiffness compared to the sample '90/10'. In general, can be concluded that the implementation of corn starch increased the strength and stiffness of the samples. The addition of metakaolin to the mixture, however, reduced the strength of the samples.

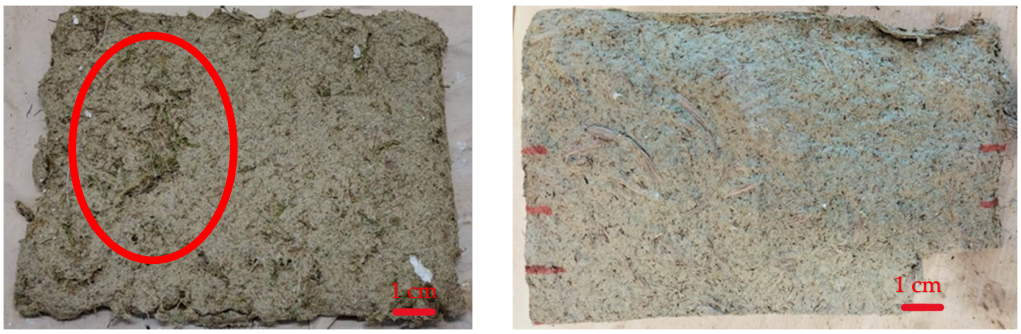
The sample '100/0-M60' was fragile and crumbled rapidly (Figure 4c). The sample '90/10-M60' (Figure A1d), with the addition of 10% cellulose and 60g of metakaolin, resulted in a self-supporting plate with a lower strength than the other samples containing cellulose. The inclusion of cellulose pulp as a matrix was necessary to achieve the desired strength by holding all the components together. The combination of moss fibers, cellulose pulp, metakaolin, and corn starch, '90/10-C15-M30' (Figure 4d), was more robust compared to the other samples. The addition of cellulose fibers and corn starch provided a plate of strength and stiffness, while the combination of corn starch and metakaolin could improve the water resistance, as suggested in the State-of-the-art section.

Apart from the assessed stiffness and strength of the samples, it could be observed that a number of samples were curved after the drying period. The samples could warp due to uncontrolled evaporation, meaning that water evaporated faster in certain areas of the sample. This phenomenon occurred with the samples solely composed of moss and cellulose, Figure 5a, and with the addition of corn starch. In contrast, the samples containing metakaolin did not warp, see Figure 5b. This can be attributed to the better water retention properties of metakaolin, resulting in a controlled and gradual evaporation process.

(**a**) (**b**)

Figure 5. (**a**) Sample '90/10' curved after drying period; (**b**) sample '90/10-M60' preserved shape after drying period.

The implementation of long fibers (moss or jute) in the substrate also had a visual influence on the samples. The sample '90/10-LM30' (Figure 6a), which contained 30 g of untreated moss fibers, could not be compacted as well as the other samples, resulting in a thicker plate. The high content of long fibers, together with the thickness of the moss fibers, may have caused this. Moreover, the fibers could not be spread homogenously because they were not blended in the mixer, resulting in high concentrations of moss fibers at some spots. Next, the sample '90/10-LM15-C15' was manufactured with a lower untreated moss fiber content and the addition of corn starch. In this case, the plate could be compressed better, making the plate thinner again. It can also be remarked that the flexibility of the plate slightly increased. The sample '90/10-LJ5' (Figure 6b), with the addition of jute fibers (thinner than the moss fibers), exhibited similar results.



(**a**) (**b**)

Figure 6. (**a**) Sample '90/10-LM30', the circle indicates a zone with a high concentration of moss fibers; (**b**) sample '90/10-LJ5'.

The samples could hypothetically exhibit enhanced performance if the moss fibers were to be uniformly distributed, minimizing interference among the fibers, removing weak spots without fibers, and achieving an improved enclosure of the fibers by the matrix. The fibers could be dispersed more evenly by optimizing the mix design and by mechanical mixing of the fibers and the matrix. The content of long fibers was decreased from 15 g (sample 90/10-LM15) to 5 g (sample '90/10-LJ5') because proper distribution proved unachievable with a high content of long fibers. This adjustment in sample composition resulted in a better distribution of the long fibers, as observed in Figure 6b. Additionally, by utilizing the vibrational mixing method already established in the concrete industry, enhancements in the distribution can be achieved [48].

4.2. Microscopic Assessment of Samples

The *Sphagnum* moss had a remarkable structure when studied under the microscope. The branches were wide open when the fiber was moistened, while they closed upon drying to conserve as much water as possible. This structure prompted the concept of applying pressure to the moist fiber to remove excess water, interlocking the moss fibers and potentially incorporating binders in between. Figure 4a shows a microscopic image of sample '100/0'. It was observed that the moss fibers were overlapped and mixed in all directions. There were voids between the individual moss fibers, which may explain the weak bonding perceived in the manual assessment. The sample '90/10' showed a structure of moss fibers combined with cellulose pulp (Figure 4b). It was noticeable that the individual moss fibers could be distinguished anymore because they were enveloped by the cellulose pulp. The voids in the structure were filled with cellulose fibers, resulting in a dense structure with a high degree of homogeneity. The larger components that can be perceived are imperfections, namely organic materials like grass, sand, and sticks. Observing sample '100/0-C15' under the microscope (Figure A2a), it can be noticed that the corn starch cannot be distinguished in the image as the entire mixture formed a whole. The moss fibers were enveloped by the corn starch, resulting in a dense and homogeneous microstructure, where the gaps revealed in the microstructure of sample '100/0' were filled. The sample '100/0-M60' (Figure 4c) illustrates the moss fibers, but this time they were enclosed by the metakaolin. The moss fibers were easily distinguishable from each other compared to the samples with cellulose or corn starch. Furthermore, the metakaolin particles were evenly spread around the plate. However, the samples with cellulose and corn starch were a coherent mixture. The sample '100/0-M60' showed two materials, where metakaolin was a filler rather than a binder, which can also be noted by visual inspection. The observation of the discussed samples under the microscope made it possible to distinguish the various components in the samples '90/10-C15' (Figure A2b), '90/10-M60' (Figure A2c), and '90/10-C15-M30', see Figure 4d.

4.3. Tensile Strength

The tensile strength between the samples varied from 0.1 to 0.71 MPa (Figure 7). Three samples were excluded from the test due to their fragility, viz., '100/0', '100/0-M60', and '90/10-M60'. Cellulose was required to obtain a strong sample that could support its own weight. When moss was combined with solely metakaolin or corn starch, the plate was not able to stand on its own. For this reason, the sample containing metakaolin was not able to be tested, and the sample with solely moss and corn starch was the weakest of the tested samples. Furthermore, the higher the amount of cellulose, the greater the tensile strength, varying from 0.24 MPa with 10% cellulose to 0.71 MPa with 50% cellulose.

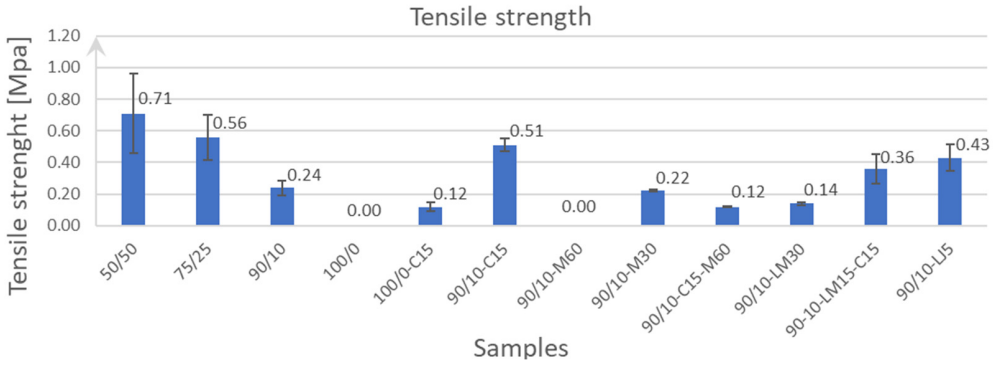


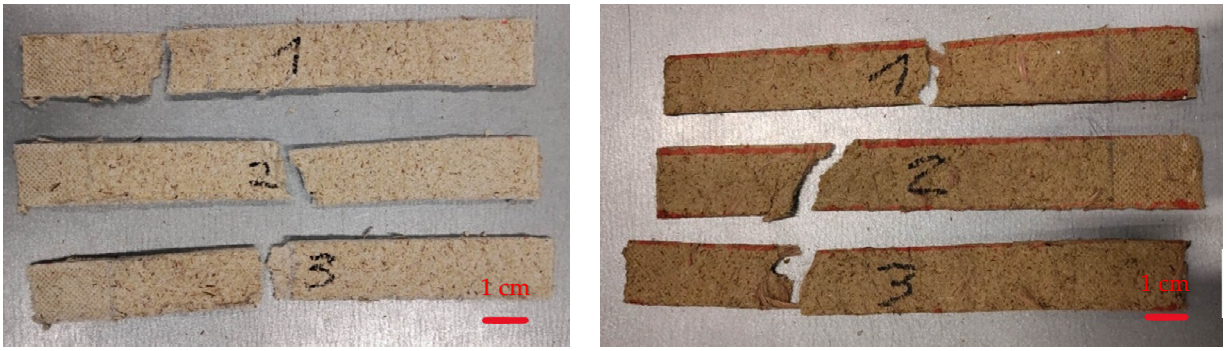
Figure 7. Average tensile strength in MPa measured for three specimens of each sample composition.

The addition of corn starch to enhance the tensile strength was effective. Adding 15 g of corn starch to the sample with 90% moss and 10% cellulose improved the tensile strength significantly from 0.24 MPa to 0.51 MPa. Furthermore, the addition of 30 g of

metakaolin to the sample of 90% moss and 10% cellulose caused a slight decrease in the tensile strength from 0.24 MPa to 0.22 MPa. In contrast, adding more metakaolin reduced the tensile strength significantly. The sample containing 60 g of metakaolin was too weak to perform the tensile test. Additionally, when corn starch, cellulose, and metakaolin were combined, the sample maintained its structural integrity but showed the weakest results of all the samples. The incorporation of long moss fibers decreased the tensile strength. The sample '90/10-LM30' reduced the tensile strength to 0.12 MPa in comparison to sample '90/10' with 0.24 MPa. Furthermore, the addition of corn starch did not improve the tensile strength. Sample '90/10–LM15-C15' had a tensile strength of 0.36 MPa compared to the 0.51 MPa tensile strength of sample '90/10–C15'. In the last sample, long jute fibers were added. In comparison with the previous sample, the addition of jute improved the tensile strength, with an increase from 0.24 MPa (sample '90/10') to 0.43 MPa (sample '90/10–LJ5'), due to the high tensile strength of the jute fibers.

Finally, it is worth mentioning that the standard deviation of the samples was notably high. This was primarily due to the wide variations in plate thickness, a consequence of the suboptimal manufacturing process. Nevertheless, the results still provide valuable insights into the tensile strength of the sample compositions.

The fracture characteristics of the materials exhibited a horizontal fracture line, which implies a brittle fracture. Furthermore, the fracture line often appeared at the edge, where the samples were held, due to the stress concentration, see Figure 8a. The fracture lines that occurred with the samples containing long fibers varied. With long fibers, the fracture followed the direction of the long fibers, which can be seen in Figure 8b. Lastly, the strain of the samples could not be measured during testing, as the test did not automatically stop and required manual intervention to terminate. However, if the measurements of sample strain are considered necessary, it can be achieved through conducting a bending test.



(**a**) (**b**)

Figure 8. (**a**) Fracture lines observed in the three samples of the '90/10-M30'sample type; (**b**) fracture lines observed in the three samples of the '90/10-LJ5' sample type.

4.4. Water Permeability

Initially, an attempt was made to measure the air permeability using 'PermeaTORR AC'. However, the porosity of the samples prevented the successful measurement of the coefficient of air permeability, since they could not be vacuumed. Alternatively, a water permeability test was set up to get a sense of the porosity and water uptake. The results of the water permeability are shown in Figure 9, which are given in seconds for each sample. The moss-only slab was characterized by a water permeability of 180 s. The permeability decreased with an increasing amount of cellulose, resulting in 377 s with 10% cellulose and 475 s with 50% cellulose. Furthermore, the addition of corn starch had a significant impact on the water permeability. The sample with moss and corn starch had a water passage time of 337 s compared to 180 s with solely moss. The addition of corn starch with the sample '90/10' had a significantly longer passage time of 1210 s. Lastly, metakaolin had the greatest impact on water permeability. The sample of solely moss and metakaolin was excluded from the test due to the brittleness mentioned previously; however, the addition

of metakaolin in combination with other binders had a significant impact on the water permeability, resulting in 2046 s and 2453 s, respectively. Sample '90/10–M60' was an outlier, with a passage time of 178 s; due to the weakness of the plate, it collapsed under the pressure of the water. It can be concluded that the addition of binders decreased the water permeability, resulting in the combination of binders in the least water-permeable sample. The structure of the samples containing long fibers was not as dense, which had an impact on the water permeability results. As a result, the samples with long fibers were more water-permeable than the samples without. Due to the non-homogeneous distribution of the fibers, the location where the test was performed had a greater impact on the results obtained. Therefore, a single test on one location will not be sufficient enough to draw a conclusion. Ultimately, the water permeability test was performed on synthetic fabric ('SF'). The water fell directly through the fabric with a passage time of 5 s, being more water-permeable than all the samples. However, the developed samples could retain water considerably better, which is essential for growing moss on them. The low porosity could be solved by providing samples of perforations, allowing airflow to pass through.

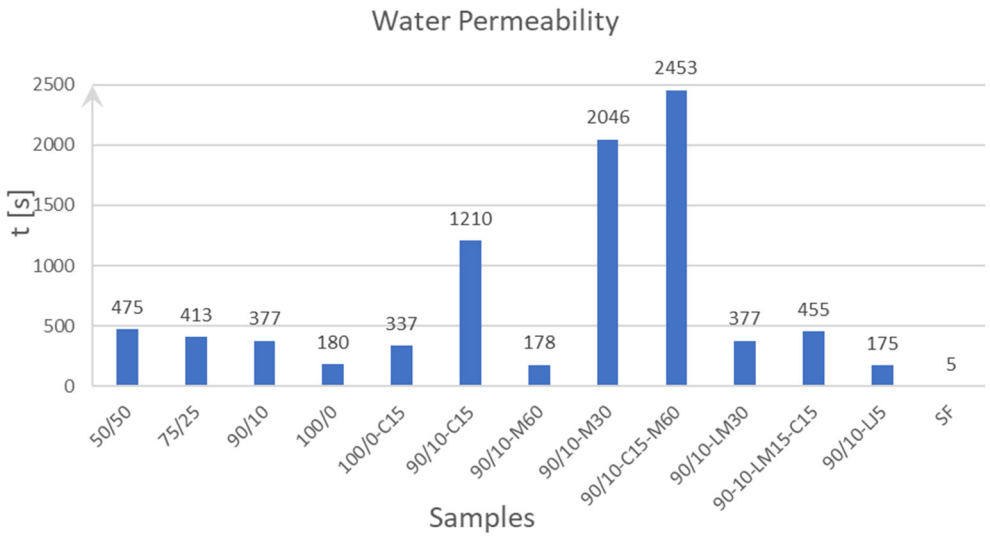


Figure 9. Single-test water permeability in s measured for the different sample compositions.

4.5. Bioreceptivity

To create optimal conditions for bioreceptivity, the samples were enveloped with small openings for ventilation and regularly moistened to maintain ideal moisture levels. Furthermore, the pH was intentionally adjusted towards acidity by applying yoghurt to stimulate moss growth. The key parameters governing bioreceptivity, such as water retention capacity and nutrient availability, were also considered. The resulting biocomposite, composed entirely of organic materials, provided a nutrient-rich environment that can stimulate moss growth. Notably, this material demonstrated enhanced water retention capabilities compared to the original synthetic microfiber cloth. Additionally, the augmented surface roughness emerged as a significant factor contributing to improved bioreceptivity, offering protection to microorganisms and against weathering effects [49,50].

The samples were observed for one month under the conditions described above, but no moss growth could be observed. However, during this period, fungal growth was observed on the samples, aligning with anticipated outcomes described in the literature. Moss growth should be monitored for a larger period in future research to obtain more accurate results.

5. Discussion

In this research, a novel biocomposite was developed utilizing moss and natural binders (e.g., cellulose, metakaolin, and corn starch). The obtained biocomposite is 100%

recyclable, contains a significant amount of moss, and is self-supporting, making it a promising bio-based and sustainable alternative to a synthetic fabric as a substrate in moss walls. Particularly, the combination of these binders together with moss to develop a complete biodegradable composite has not yet been explored in other studies. This study also presents an innovative and detailed production process of the biocomposite, inspired by paper and nori manufacturing, and which was established using a trial-and-error approach on a lab scale. In the literature, there are also a wide range of papers investigating the production process of biocomposites. Nevertheless, it can be observed that the techniques that are mainly applied, extrusion and hot-pressing, require high temperatures, resulting in adverse environmental impacts [43,44,51]. The process established in this paper is a more energy-efficient, straightforward, and environmentally friendly alternative.

The tensile strengths measured for the samples in this study varied from 0.1 to 0.7 MPa. These values are lower than the value reported by Zhang et al. (2019), which varied from 1 to 1.5 MPa from plug-in trays, where they tested the physical and mechanical properties of commercially available biodegradable plug-in trays made from peat moss and wooden fibers [32]. Furthermore, Méité et al. (2022) investigated the addition of metakaolin in corn-starch-based bioplastics. The tensile strength of those biocomposites varied from 5.5 to 8.1 MPa [28]. These reported values contradict the measurements of this study, where the addition of metakaolin decreased the tensile strength. This could have several explanations. First of all, the amount of metakaolin added was much higher in this study and varied from 20 to 33% compared to from 0 to 15%, whereas a value of 33% showed significantly lower results. Moreover, metakaolin could be activated with calcium hydroxide ($Ca(OH)_2$), such as in concrete, in order to improve the strength; however, chemical processes were avoided in this study [38]. Furthermore, the addition of corn starch significantly improved the tensile strength of the samples, with an increase from 0.24 to 0.51 MPa; however, the addition of corn starch was limited to 15 g per plate, which is a rather low amount. Therefore, the addition of corn starch could be increased to up to 30 g or 60 g per plate. Furthermore, it is important to note that the tensile strength tests gave a rough idea of the strength of the plates as well as a comparison between the different contents of the plates. Moreover, in order to provide a more realistic evaluation of the strength of the plates, it is essential to conduct strength measurements under wet conditions.

The water permeability test performed in this study gave a general idea of the difference in the permeability and uptake of water of the different samples. It was remarkable that the incorporation of the binders decreased the water permeability of the samples, with metakaolin having the most pronounced impact. The combination of metakaolin and corn starch resulted in the lowest water permeability, with a time passage of 2453 s. This outcome is confirmed by the finding of Méité et al. (2022), in which the addition of metakaolin in a starch-based bioplastic decreased the water permeability [28]. Their results indicated that the coefficient of permeability decreased from 6.15 to 3.43×10^{-11} g/msPa with increasing levels of metakaolin (from 0 to 15%). It can be concluded that the combination of starch and metakaolin is essential in the design for enhanced water uptake and resistance, enabling the samples to be more resilient against weathering.

The results obtained during the observation of moss growth were compared with findings from other publications. This comparative analysis aimed to establish a comprehensive understanding of moss growth and to draw comparisons with results documented in the existing literature. Perini et al. (2022) investigated moss cultivation and growth chamber tests [9]. The procedure followed in this paper was very similar to the described 'solid culture', in which a moss paste is manufactured by mixing moss and agar blended in a mixer. The results showed that after five months of incubation, all the tested species managed to grow on the testers. Moss growth could only be noticed as of two months. It may be further noted that both the samples were faced with a fungal attack. Udawattha et al. (2018) investigated the growth of moss on walling materials such as concrete and bricks [45]. In this case, the culture media was composed of moss and butter milk mixed together. It can be concluded that fungal propagation is very common since it is favored by

a moist environment. Additionally, moss growth should be monitored for a longer period in future research to obtain accurate results.

In conclusion, the incorporation of the three binders has proven crucial in achieving a plate that aligns with the specified criteria. Cellulose and corn starch have demonstrated the capacity to enhance the plate's strength, while metakaolin has effectively reduced the water permeability. Nevertheless, it is important to note that this study primarily concentrated on assessing the properties and combination of diverse binders. Further exploration is needed to obtain the optimal combination of binders.

6. Conclusions and Future Research

Biocomposites have attracted the interest of researchers and led to research into natural fibers and biodegradable polymers with the aim of developing viable and sustainable alternatives to petroleum-based products. However, research on fully biodegradable biocomposites is at an early stage, and the application of moss fibers in this context is limited-to-non-existent, focusing only on creating insulation plates. This research aimed to provide additional insights regarding the development of fully biodegradable biocomposites containing a substantial proportion of recycled moss fibers. In doing so, it makes a valuable contribution to the documentation of nature-based solutions in the state of the art.

Currently, within moss wall air purification systems, a synthetic fabric is used as a substrate for moss. This raises concern about the sustainability of this system, as it is associated with a negative impact on the environment and biota. Therefore, the primary objective of this preliminary study was to explore a nature-based alternative to address this concern. The initial step involved assessing the feasibility of developing a moss-based, fully biodegradable biocomposite designed to possess structural integrity and the ability to support its self-weight.

Various combinations of natural binders and fibers were experimented with in a lab environment to achieve this goal. The addition of cellulose was found to be crucial to obtaining a sample with the capacity to bear its self-weight, with the optimal ratio of 90/10 (moss/cellulose) yielding robust results and containing a significant amount of moss fibers. The addition of corn starch in combination with cellulose improved the tensile strength significantly and decreased the water permeability. In contrast, the incorporation of metakaolin reduced the tensile strength of the samples and demonstrated the most pronounced decrease in water permeability. The combination of moss fibers, cellulose, corn starch, and metakaolin yielded a sample with structural integrity and that was capable of bearing its self-weight, with the lowest water permeability and best water retention. Compared to the synthetic fabric, which was very permeable, the samples were less permeable and could retain water better, making them suitable for supporting moss growth. Because bioreceptivity is an essential parameter in evaluating the applicability of developed materials as substrates for moss wall air purification systems, the monitoring of moss growth should be investigated for a longer period in future research. Furthermore, this study explored shape variations using a 3D-printed mold, as the plate relied on its shape to provide stiffness.

This research highlights the essentiality of the three binders; cellulose and corn starch improved the strength, while metakaolin enhanced the water resistance. Future investigation is needed to determine the optimal ratios, suggesting increasing the amount of corn starch and reducing the amount of metakaolin to obtain samples with a high tensile strength and water retention. Additionally, future research is needed to scale up the plate size and further investigate the effect of moss growth on the plate and for determining the ideal form, timing, and methodology for optimal moss growth. A crucial aspect for future research is the assessment of the durability of the samples. Key parameters, including resistance to weathering, cyclic loading and biodegradation time, can be determined by testing the samples in a climatic chamber, which is available at the University of Antwerp. While currently in the conceptual stage, if developed further, the biocomposite has the

potential to replace not only the substrate but also other essential elements. In further steps, the biocomposite could serve as a bio-based alternative building material.

Author Contributions: Conceptualization, J.B., R.A.G., L.V.d.B., B.B. and A.D.B.; methodology, J.B., R.A.G., L.V.d.B., B.B. and A.D.B.; software, R.A.G. and L.V.d.B.; validation, J.B., R.A.G. and L.V.d.B.; formal analysis, R.A.G. and L.V.d.B.; investigation, R.A.G. and L.V.d.B.; resources, J.B.; data curation, J.B.; writing—original draft preparation, R.A.G. and L.V.d.B.; writing—review and editing, J.B., B.B. and A.D.B.; visualization, R.A.G. and L.V.d.B.; supervision, J.B., B.B. and A.D.B.; project administration, J.B., B.B. and A.D.B.; funding acquisition, A.A., B.B. and J.B. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the University of Antwerp Valorisation Office under the BlueApp PoC Call, which resulted from the Flemish Resilience Recovery Plan, grant number PS ID 48157—OZ 9197.

Institutional Review Board Statement: Not applicable.

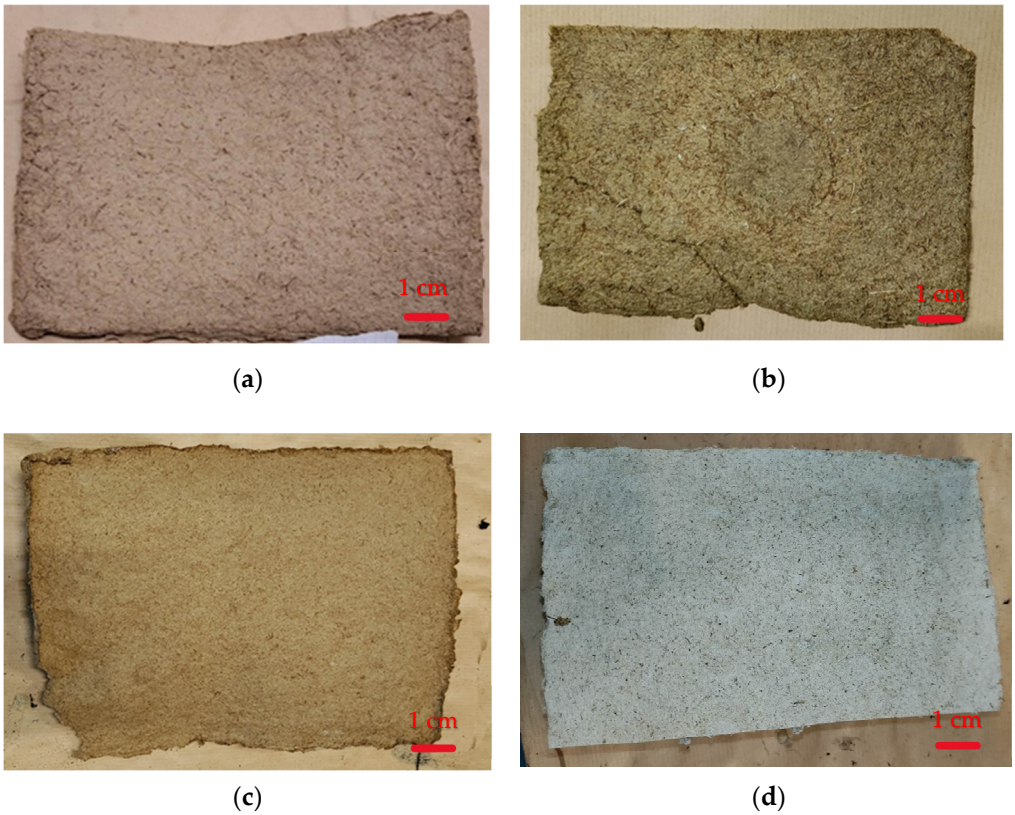
Informed Consent Statement: Not applicable.

Data Availability Statement: Data are contained within the article.

Acknowledgments: The authors would like to thank the infrastructure department of the University of Antwerp for facilitating the relocation of the moss wall to the campus.

Conflicts of Interest: There are no conflict of interest to report.

Appendix A



(**a**) (**b**)

(**c**) (**d**)

Figure A1. Overview of the samples with different combinations and proportions between fibers and binders. The composition of the samples is listed in Table 1: (**a**) sample '50/50'; (**b**) sample '100/0-C15'; (**c**) sample '90/10-C15'; (**d**) sample '90/10-M60'.

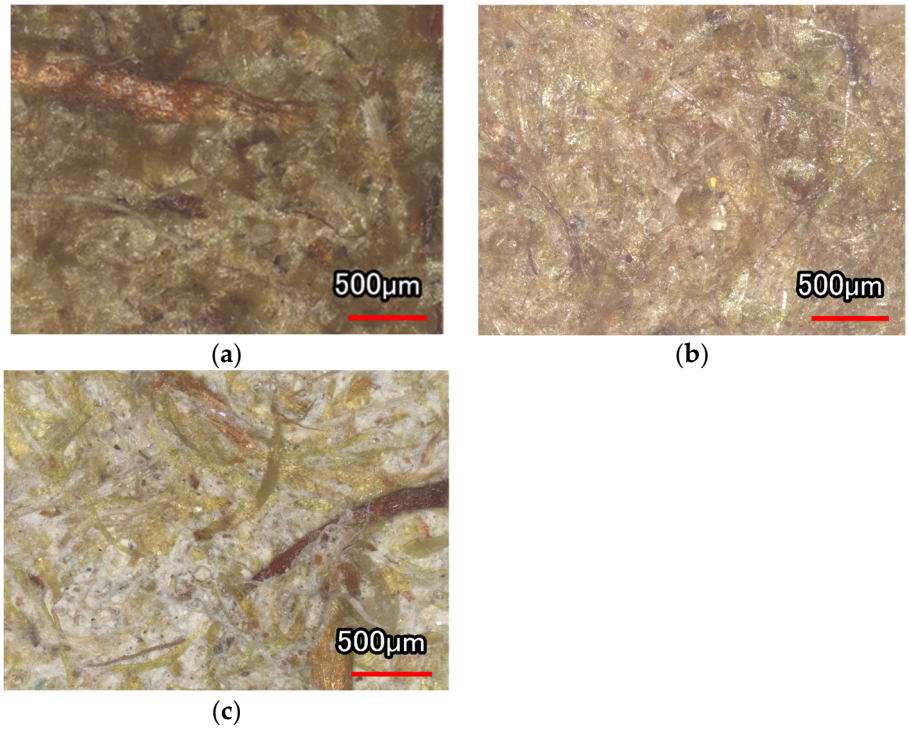


Figure A2. Microscopic images of samples up-scaled 90×: (**a**) sample '100/0-C15'; (**b**) sample '90/10-C15'; (**c**) sample '90/10-M60'.

References

1. WHO. Air Quality and Health. Available online: https://www.who.int/teams/environment-climate-change-and-health/air-quality-and-health/sectoral-interventions/ambient-air-pollution/health-risks (accessed on 29 January 2023).
2. Ysebaert, T.; Koch, K.; Samson, R.; Denys, S. Green Walls for Mitigating Urban Particulate Matter Pollution—A Review. *Urban For. Urban Green.* **2021**, *59*, 127014. [CrossRef]
3. European Environment Agency. Air Pollution: How It Affects Our Health. Available online: https://www.eea.europa.eu/themes/air/health-impacts-of-air-pollution#:~:text=Both%2520short%252D%2520and%2520long%252Dterm,asthma%2520and%2520lower%2520respiratory%2520infections (accessed on 12 January 2023).
4. European Environment Agency. Premature Deaths Due to Exposure to Fine Particulate Matter in Europe. Available online: https://www.eea.europa.eu/ims/health-impacts-of-exposure-to (accessed on 29 January 2023).
5. EPA Health and Environmental Effects of Particulate Matter (PM). Available online: https://www.epa.gov/pm-pollution/health-and-environmental-effects-particulate-matter-pm (accessed on 29 January 2023).
6. Ji, X.; Huang, J.; Teng, L.; Li, S.; Li, X.; Cai, W.; Chen, Z.; Lai, Y. Advances in Particulate Matter Filtration: Materials, Performance, and Application. *Green Energy Environ.* **2023**, *8*, 673–697. [CrossRef]
7. Wade, J.; Farrauto, R.J. 12—Controlling Emissions of Pollutants in Urban Areas. In *Metropolitan Sustainability*; Zeman, F., Ed.; Woodhead Publishing Series in Energy; Woodhead Publishing: Cambridge, UK, 2012; pp. 260–291, ISBN 978-0-85709-046-1.
8. Blocken, B.; Vervoort, R.; van Hooff, T. Reduction of Outdoor Particulate Matter Concentrations by Local Removal in Semi-Enclosed Parking Garages: A Preliminary Case Study for Eindhoven City Center. *J. Wind. Eng. Ind. Aerodyn.* **2016**, *159*, 80–98. [CrossRef]
9. Perini, K.; Castellari, P.; Gisotti, D.; Giachetta, A.; Turcato, C.; Roccotiello, E. MosSkin: A Moss-Based Lightweight Building System. *Build. Environ.* **2022**, *221*, 109283. [CrossRef]
10. European Commission. Ceramic Honeycomb Air Filters Could Cut City Pollution. Available online: https://ec.europa.eu/research-and-innovation/en/horizon-magazine/ceramic-honeycomb-air-filters-could-cut-city-pollution (accessed on 29 January 2023).
11. BESIX. BESIX: Proefproject "Clean Air" Tegen Fijnstof. Available online: https://press.besix.com/besix-proefproject-clean-air-tegen-fijnstof (accessed on 29 January 2023).
12. Green City Solutions the Citytree. Available online: https://greencitysolutions.de/en/citytree/ (accessed on 29 January 2023).
13. Whitton, J. *Plant Biodiversity, Overview*, 2nd ed.; Levin, S.A., Ed.; Academic Press: Cambridge, MA, USA, 2013.
14. EnGreDi Mosses. Available online: https://www.engredi.be/mosses (accessed on 31 January 2023).
15. European Commission. Using Moss to Measure Air Pollution. Available online: https://ec.europa.eu/newsroom/horizon2020/items/15130#:~:text=%25E2%2580%259CMosses%2520lack%2520a%2520root%2520system,and%2520gaseous%252C%25E2%2580%259D%2520he%2520adds (accessed on 12 January 2023).

16. BESIX BESIX Clean Air Neemt Een Groeispurt, Net Als Haar Mossen. Available online: https://www.besix.com/nl/news/besix-clean-air-neemt-een-groeispurt-net-als-haar-mossen (accessed on 29 January 2023).
17. United States Environmental Protection Agency. Microplastics Research. Available online: https://www.epa.gov/water-research/microplastics-research (accessed on 15 March 2023).
18. Jahandari, A. Microplastics in the Urban Atmosphere: Sources, Occurrences, Distribution, and Potential Health Implications. *J. Hazard. Mater. Adv.* **2023**, *12*, 100346. [CrossRef]
19. Nguyen, M.-K.; Lin, C.; Nguyen, H.-L.; Le, V.-R.; KL, P.; Singh, J.; Chang, S.W.; Um, M.-J.; Nguyen, D.D. Emergence of Microplastics in the Aquatic Ecosystem and Their Potential Effects on Health Risks: The Insights into Vietnam. *J. Environ. Manag.* **2023**, *344*, 118499. [CrossRef]
20. de Smet, D.; Goethals, F.; Demedts, B.; Uyttendaele, W.; Vanneste, M. Bio-Based Textile Coatings and Composites. In *Biobased Products and Industries*; Elsevier Inc.: Amsterdam, The Netherlands, 2020; pp. 357–402, ISBN 9780128201534.
21. Vitola, L.; Gendelis, S.; Sinka, M.; Pundiene, I.; Bajare, D. Assessment of Plant Origin By-Products as Lightweight Aggregates for Bio-Composite Bounded by Starch Binder. *Energies* **2022**, *15*, 5330. [CrossRef]
22. Faruk, O.; Bledzki, A.K.; Fink, H.P.; Sain, M. Biocomposites Reinforced with Natural Fibers: 2000–2010. *Prog. Polym. Sci.* **2012**, *37*, 1552–1596. [CrossRef]
23. Madurwar, M.V.; Ralegaonkar, R.V.; Mandavgane, S.A. Application of Agro-Waste for Sustainable Construction Materials: A Review. *Constr. Build. Mater.* **2013**, *38*, 872–878. [CrossRef]
24. Kain, G.; Idam, F.; Tonini, S.; Wimmer, A. Torfmoos (Sphagnum)—Historisches Erfahrungswissen Und Neue Einsatzmöglichkeiten Für Ein Naturprodukt. *Bauphysik* **2019**, *41*, 199–204. [CrossRef]
25. Morandini, M.C.; Kain, G.; Eckardt, J.; Petutschnigg, A.; Tippner, J. Physical-Mechanical Properties of Peat Moss (Sphagnum) Insulation Panels with Bio-Based Adhesives. *Materials* **2022**, *15*, 3299. [CrossRef]
26. Bakatovich, A.; Gaspar, F. Composite Material for Thermal Insulation Based on Moss Raw Material. *Constr. Build. Mater.* **2019**, *228*, 116699. [CrossRef]
27. Marichelvam, M.K.; Jawaid, M.; Asim, M. Corn and Rice Starch-Based Bio-Plastics as Alternative Packaging Materials. *Fibers* **2019**, *7*, 32. [CrossRef]
28. Méité, N.; Konan, L.K.; Tognonvi, M.T.; Oyetola, S. Effect of Metakaolin Content on Mechanical and Water Barrier Properties of Cassava Starch Films. *S. Afr. J. Chem. Eng.* **2022**, *40*, 186–194. [CrossRef]
29. Lorente-Ayza, M.-M.; Sánchez, E.; Sanz, V.; Mestre, S. Influence of Starch Content on the Properties of Low-Cost Microfiltration Ceramic Membranes. *Ceram. Int.* **2015**, *41*, 13064–13073. [CrossRef]
30. Ferrari, F.; Striani, R.; Fico, D.; Alam, M.M.; Greco, A.; Esposito Corcione, C. An Overview on Wood Waste Valorization as Biopolymers and Biocomposites: Definition, Classification, Production, Properties and Applications. *Polymers* **2022**, *14*, 5519. [CrossRef]
31. Fuentes, R.A.; Berthe, J.A.; Barbosa, S.E.; Castillo, L.A. Development of Biodegradable Pots from Different Agroindustrial Wastes and Byproducts. *Sustain. Mater. Technol.* **2021**, *30*, e00338. [CrossRef]
32. Zhang, X.; Wang, C.; Chen, Y. Properties of Selected Biodegradable Seedling Plug-Trays. *Sci. Hortic.* **2019**, *249*, 177–184. [CrossRef]
33. Kain, G.; Morandini, M.; Stamminger, A.; Granig, T.; Tudor, E.M.; Schnabel, T.; Petutschnigg, A. Production and Physical–Mechanical Characterization of Peat Moss (Sphagnum) Insulation Panels. *Materials* **2021**, *14*, 6601. [CrossRef]
34. Ramesh, M.; Palanikumar, K.; Reddy, K.H. Plant Fibre Based Bio-Composites: Sustainable and Renewable Green Materials. *Renew. Sustain. Energy Rev.* **2017**, *79*, 558–584. [CrossRef]
35. Bakshi, P.S.; Selvakumar, D.; Kadirvelu, K.; Kumar, N.S. Chitosan as an Environment Friendly Biomaterial—A Review on Recent Modifications and Applications. *Int. J. Biol. Macromol.* **2020**, *150*, 1072–1083. [CrossRef]
36. Rubino, C.; Aracil, M.B.; Gisbert-Payá, J.; Liuzzi, S.; Stefanizzi, P.; Cantó, M.Z.; Martellotta, F. Composite Eco-Friendly Sound Absorbing Materials Made of Recycled Textilewaste and Biopolymers. *Materials* **2019**, *12*, 4020. [CrossRef] [PubMed]
37. Khatib, J.M.; Baalbaki, O.; ElKordi, A.A. 15—Metakaolin. In *Waste and Supplementary Cementitious Materials in Concrete*; Siddique, R., Cachim, P., Eds.; Woodhead Publishing Series in Civil and Structural Engineering; Woodhead Publishing: Cambridge, UK, 2018; pp. 493–511, ISBN 978-0-08-102156-9.
38. Sabir, B.B.; Wild, S.; Bai, J. Metakaolin and Calcined Clays as Pozzolans for Concrete: A Review. *Cem. Concr. Compos.* **2001**, *23*, 441–454. [CrossRef]
39. Pundiene, I.; Vitola, L.; Pranckeviciene, J.; Bajare, D. Hemp Shive-Based Bio-Composites Bounded by Potato Starch Binder: The Roles of Aggregate Particle Size and Aspect Ratio. *J. Ecol. Eng.* **2022**, *23*, 220–234. [CrossRef]
40. Mohamad Amini, M.H.; Hashim, R.; Sulaiman, N.S.; Sulaiman, O.; Selamat, M.E. Glutardialdehyde Modified Starch from Waste Oil Palm Trunks as a Binder for Wood Composite Making. *Int. J. Adhes. Adhes.* **2021**, *104*, 102757. [CrossRef]
41. Mawardi, I.; Aprilia, S.; Faisal, M.; Rizal, S. Investigation of Thermal Conductivity and Physical Properties of Oil Palm Trunks/Ramie Fiber Reinforced Biopolymer Hybrid Composites as Building Bio-Insulation. *Mater. Today Proc.* **2022**, *60*, 373–377. [CrossRef]
42. Fatima, A.; Qayyum, N.; Zaid, M.; Rafat, Y.; Zain Khan, M.; Ali, R.; Salem, B. Feasibility of Manufacturing Sustainable Bio-Composites from Agricultural Waste. *Mater. Today Proc.* **2021**, *47*, 7136–7139. [CrossRef]
43. Edebali, S. 15—Methods of Engineering of Biopolymers and Biocomposites. In *Advanced Green Materials*; Ahmed, S., Ed.; Woodhead Publishing in Materials; Woodhead Publishing: Cambridge, UK, 2021; pp. 351–357, ISBN 978-0-12-819988-6.

44. Viretto, A.; Gontard, N.; Angellier-Coussy, H. Urban Parks and Gardens Green Waste: A Valuable Resource for the Production of Fillers for Biocomposites Applications. *Waste Manag.* **2021**, *120*, 538–548. [CrossRef]
45. Udawattha, C.; Galkanda, H.; Ariyarathne, I.S.; Jayasinghe, G.Y.; Halwatura, R. Mold Growth and Moss Growth on Tropical Walls. *Build. Environ.* **2018**, *137*, 268–279. [CrossRef]
46. Flashforge Flashforge Guider II. Available online: https://www.flashforge.com/product-detail/10 (accessed on 23 May 2023).
47. Leitch, A. Recycled Cardboard Molding. Available online: https://www.thingiverse.com/thing:3912997 (accessed on 23 May 2023).
48. Xiong, G.; Wang, C.; Zhou, S.; Zheng, Y. Study on Dispersion Uniformity and Performance Improvement of Steel Fibre Reinforced Lightweight Aggregate Concrete by Vibrational Mixing. *Case Stud. Constr. Mater.* **2022**, *16*, e01093. [CrossRef]
49. Veeger, M.; Prieto, A.; Ottelé, M. Exploring the Possibility of Using Bioreceptive Concrete in Building Façades. *J. Facade Des. Eng.* **2021**, *9*, 73–86. [CrossRef]
50. Veeger, M.; Ottelé, M.; Prieto, A. Making Bioreceptive Concrete: Formulation and Testing of Bioreceptive Concrete Mixtures. *J. Build. Eng.* **2021**, *44*, 102545. [CrossRef]
51. Nguyen, D.M.; Grillet, A.-C.; Bui, Q.-B.; Diep, T.M.H.; Woloszyn, M. Building Bio-Insulation Materials Based on Bamboo Powder and Bio-Binders. *Constr. Build. Mater.* **2018**, *186*, 686–698. [CrossRef]