



Article Improving the Behaviour of Green Concrete Geopolymers Using Different HEMP Preservation Conditions (Fresh and Wet)

Mª Paz Sáez-Pérez ^{1,*}, Jorge Alberto Durán-Suárez ² and Joao Castro-Gomes ³

- ¹ Building Constructions Department, Advanced Technical School for Building Engineering, University of Granada, 18071 Granada, Spain
- ² Sculpture Department, Faculty of Fine Arts, University of Granada, 18071 Granada, Spain
- ³ Department of Civil Engineering and Architecture, University of Beira Interior, 6201-001 Covilhã, Portugal
 - * Correspondence: mpsaez@ugr.es

Abstract: This paper evaluates a type of geopolymer concrete that uses hemp fibres as a natural aggregate due to the various advantages offered by these woody materials. These advantages include ease of cultivation and processing and their use in the essential structure of concretes used for green construction purposes. The sampling study was prepared using an environmentally friendly inorganic binder, based on geopolymerization reactions (Si-Na). The improvement in the hemp aggregate using two different preservation methods (fresh and wet) was assessed. The type of conservation enables anaerobic reactions to take place in the structure of the hemp, in such a way as to modify the proportions of the organic compounds contained in the hemp and the morphology of the fibres. It also encourages the proliferation of cellulose nanofibrils (CNC), which enhance the mechanical results, improving plasticity and thixotropy. The hempcrete studied in this paper could be a good alternative material for sustainable, environmentally friendly construction, as much less CO_2 is emitted during the production process in comparison with conventional concrete. Using wet-preserved hemp means that less water must be added to the mix during preparation of the concrete. This also helps reduce production costs, and by extension, the cost of the final product.

Keywords: sustainable materials; concrete geopolymer; fresh and wet preservation; hemp fibres; circular economy

1. Introduction

One of the basic principles of the circular economy in the building materials sector is that new solutions and alternative proposals must be implemented to eliminate the negative impacts on the environment of certain building materials [1–3]. Normally, these tend to involve high energy consumption and non-renewable resources [4–8]. One of the clearest examples are products made of Portland cement, whose manufacturing process involves high CO_2 emissions [9]. New research has demonstrated that one of the best solutions involves using geopolymer compounds (in this case made of alkalis and strengthened with fibres) together with more sustainable new technologies [10,11].

Nowadays, given the increase in competition in the building materials market, the need to develop environmentally friendly products is becoming the main focus of the latest trends in design [12].

By definition [13,14], hempcrete, a concrete prepared with lime and hemp, is a light concrete which uses hemp hurds or shivs (waste products from hemp fibre production) as an aggregate, together with a lime-based binder, which is more compatible than cement [15]. The high proportion of materials of biological origin means that hemp lime is a net absorber of carbon dioxide with an essentially sustainable production system [16,17].

Research confirms the strong impact that the compositional and structural conditions (hemp–lime ratio, proportion of binder, porous structure [18], disposition of the fibres [19])



Citation: Sáez-Pérez, M.P.; Durán-Suárez, J.A.; Castro-Gomes, J. Improving the Behaviour of Green Concrete Geopolymers Using Different HEMP Preservation Conditions (Fresh and Wet). *Minerals* 2022, 12, 1530. https://doi.org/ 10.3390/min12121530

Academic Editors: Fernando Pelisser and Dachamir Hotza

Received: 9 October 2022 Accepted: 25 November 2022 Published: 29 November 2022

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). can have on the hempcrete's main properties, as can the way it is placed in the building and other manufacturing conditions [13]. Researchers have also found that hemp concrete has low density [20] and good thermal and acoustic insulation properties [21,22] due to its porosity and low density. It is also known for its good hygrothermal properties [23] and its high water-vapour permeability and sorption properties [23–25]. This enables it to passively regulate the humidity in a built environment, in that it can absorb relative humidity or indeed return it to the environment, in the event of any excess or lack in the comfort ambience of the room [26,27]. According to [28], it is precisely hempcrete's absorbent power that enables it to moderate the daily variations in relative humidity and guarantee the good quality of indoor air. It also shows great capacity to resist fire, mould, and fungi [14] and is known to be a non-allergenic material, as no allergic reactions are related to its use. It can therefore be viewed as a material that respects the health of living creatures [29]. Finally, it has an excellent useful life and low maintenance costs [30]. From the point of view of durability, [15] confirmed that the binder is responsible for the resistance of hempcrete during freeze/thaw cycles, exposure to salt and biodegradation.

For this reason, building materials manufacturers are increasingly replacing cement materials with geopolymers [31,32]. In addition, in recent years, there has been growing interest in the use of natural fibres as a substitute for synthetic fibres [3,8,33]. This option is usually considered to be positive from an environmental perspective because it reduces CO_2 emissions significantly [34–37]. Several studies [37–40] confirm that these natural fibre compounds are suitable for use in a range of different building materials because they have better intrinsic properties (mechanical strength, thermal resistance, low thermal conductivity, etc.), in addition to other positive characteristics that could improve their behaviour as building materials and during their manufacturing process [41].

Also, in this case, the most important advantage is that low temperatures and minimum amounts of water are used during the manufacturing process [3,42–45]. Another benefit of building materials made with natural fibres is that they have lower production costs than those made with synthetic fibres [3,8,38]. Nevertheless, there are also certain disadvantages.

The most obvious disadvantages are low compressive strength and elastic modulus [20], the loss of properties over time due to the problems of biodegradation by UV rays identified in geopolymers made with natural fibres [46], and the dissolution or precipitation of new compounds in the mixes [23]. As a highly heterogeneous and anisotropic material [47], when it is exposed to outdoor climate conditions, dimensional variations [48] and changes in shape, which can also affect its hygrothermal behaviour [49], can appear. Due to its hygroscopic nature, it is especially sensitive to variations in humidity, which cause changes in the volume of its fibres.

The use of natural fibres of this kind has shown how useful they can be in building materials in which the most important characteristics are mechanical strength and thermal and acoustic properties, with similar results to other materials with reinforcement fibres [7,50,51]. Other studies show that adding hemp fibres during the manufacture of geopolymer composites improved the mechanical properties as compared to a geopolymer without hemp [52].

Other authors [53] confirm that when hemp fibres are added to the concrete mix, their extractive molecules spread throughout the water in the mix, slowing down the kinetics of the hydration of the binder. This could alter the properties of the materials; in particular, it could reduce their mechanical resistance [54–56]. Certain mechanisms of interaction between plant molecules and mineral phases have been identified that could explain this result.

Moreover, the addition of hemp fibres changed the structural failure classification from brittle to quasi-ductile. The durability tests on hempcrete blocks and hempcrete render showed that the elements maintained their good performance in terms of mechanical properties and permeability to water vapour [57]. Hydraulic binders also improved mechanical properties and resistance to frost [58].

Finally, several studies [3,59–61] have demonstrated that industrial hemp fibre has great potential for use in building materials and is recognized as one of the most resistant, most rigid natural fibres available worldwide. In order to limit the effects of the hemp on the hydration of the binders [62–65], other authors propose adapting the formulation of the binders and their additives [66–69].

These issues are resolved and the final performance is improved using pre-treatments [70–77]. In recent years, fibre plant cultivation has increased all over the world [78,79]. Looking specifically at hemp, this is due to the fact that it does not contain herbicides and is resistant to insects and disease. Various different parts of the plant are valued positively, and it has a simpler recycling process than other plant types. As has been pointed out in [80], each tonne of hemp cultivated is capable of reabsorbing up to 1800 kg of CO₂.

Other important issues include the selection of the raw material and the processing procedure, in which the fibres that contain leaves, stems, and cores offer special advantages, as several authors have confirmed [75,81,82]. In regards to materials made of hemp fibres, cultivation and harvesting conditions are of great importance. Numerous studies [83–86] confirm that these processes cause enormous variability in the conditions of the natural fibres and in the properties of the compound materials, affecting chemical composition (proportion of hemicellulose, lignin, and cellulose) and structural features (shape, size, etc.).

If we turn to the effects of the chemical composition of the fibres, it is well-known that the improvement that cellulose nanocrystals have brought about in different sectors such as energy, water purification, the automotive industry, biomedicine, and biocomposites [87–89] is due to their excellent chemical, mechanical, and thermal properties along with other benefits such as non-toxicity, surface functionality, easy modification, and sustainability.

Furthermore, several recent studies have highlighted that the generation of cellulose nanocrystals from the most common fibres improves the behaviour of certain building materials [90–93]. In the specific case of hemp fibres, [94–96], a review of the literature confirms that the chemical pre-treatment of the fibres shows the great potential that the modification of nanosilica and CNCs in geopolymer materials has for improving their mechanical properties. These improvements are very useful but can also be a source of problems if not used properly.

As for the conditions in which the fibres are preserved and how this affects their behaviour, several studies [97,98] indicate that the climatic conditions entail certain risks. For this reason, other studies [99–101] propose simplifying the harvesting process to try to reduce the problems caused by the weather conditions at harvest time. Recently, a new harvesting technique has been developed that is unaffected by climate conditions. This technique involves a two-stage process in which the hemp is harvested and then preserved in wet storage in anaerobic conditions [102,103], during which natural fermentation takes place. This causes changes in the content of the hemp (proportional composition of the different elements) while maintaining the positive qualities of the fibres [82,104].

In this paper, the behaviour of geopolymer hempcrete with varying compositions in different states of preservation was studied. The main goal was to find out whether the preservation of the hemp fibres in different forms during the storage process affects their behaviour when used in building materials.

Our second objective was to demonstrate that the use of this type of green material can reduce costs, as it has better qualities and can be stored in a humid state, reducing costs when growing conditions are not adequate or when it cannot be grown in the native area. All these factors involve the optimization of natural resources. To this end, the environmental sustainability assessment can identify the impact of different farming practices (choice of harvest time, cultivar, conservation conditions, etc.) on industrial hemp cultivation to produce fibres that can be used in building materials. This research seeks to demonstrate that many products can have circular value if the conservation conditions are studied and possible new uses are identified. It is also important to remember that circular economy models can help provide a huge competitive advantage, not only in terms of their objectives but also because they produce more value from available resources than traditional production and consumption models [105,106].

2. Materials and Methods

In this research, the samples were prepared with an organic hemp binder and inorganic materials. The materials used as the organic binder were from the Fedora 17 variety of hemp. It had two different states of conservation (freshly harvested hemp (H) and hemp preserved wet in anaerobic conditions for six months (A)). The inorganic materials were clay, sodium silicate, glass powder, and sodium hydroxide. In both cases, after harvesting, the hemp was stored in hermetically sealed plastic containers. Both types of hemp (recently harvested and wet-preserved shredded hemp) were frozen until the different experiments required for this research were conducted.

In order to obtain the shiv, the hemp plant is subjected to a mechanical grinding process (forage harvester) in which the stem is separated from the rest of the plant and later cut up into pieces up to 50 mm long. It is then pressed and packaged with silage films to remove any oxygen and prevent the degradation of the cellulose during the wet storage period.

When the freshly harvested hemp is compared with the hemp preserved in anaerobic conditions for six months, several changes can be observed in the pH and in the content of the metabolic products due to the metabolic activity of anaerobic bacteria (Table 1).

Metabolic Products	Freshly Harvested Hemp (0 Months) H (%)	Anaerobic Wet-preserved Hemp (6 Months) A (%)				
Cellulose	57.25	59.54				
Hemicellulose	13.25	11.40				
Lignin	9.84	8.51				
Alcohols	1.29	1.03				
Lactic acid	0.76	0.6				
Total acids	1.34	2.32				
pН	7.62	5.87				

Table 1. Proportion of structural polymers, content of metabolic products and pH in freshly-harvested (H) and anaerobic wet-preserved hemp (A). Data taken from [107].

In particular, we found that in the wet-preserved samples (A), there was a decrease in the percentage of lignin (13.5%) and hemicellulose (14%), and an increase in cellulose compound (5%) as compared to the freshly harvested samples (H). There was also a reduction in alcohols (20%) and lactic acid (21%). Total acid content increased sharply (73%).

Finally, we observed that after the 6-month preservation period (A), there was a 23% reduction in pH, a finding that confirms, in line with [38], that wet preservation increases the total acidity of hemp.

Considering that a higher content of fine particles and/or dust in the fibrous material can have a negative effect on the strength properties of the manufactured materials [68,69], a detailed analysis of the hemp particles of the material (with both ways of preservation, H-A) was performed using the Fibre Shape image analysis system (IST AG, Vilters, Ebnat-Kappel, Switzerland) [108]. This analysis, performed by the Leibniz Institute for Agricultural Engineering and Bioeconomics (ATB, Potsdam, Germany) [107], provides insight into the frequency distribution of fibre width (Figure 1).

As for fibre shape, Figure 1 shows that the thinnest fibres (smallest widths) became even thinner, and the thickest fibres (greatest width) became even thicker. In this respect, over an approximate range of 0.136 to 0.251, a fibre thickening of almost 10 units can be observed from freshly harvested hemp to anaerobic wet-preserved hemp, while in the range of 0.251–2.895 it was almost double. All of this is due to the process of wet storage for six months and the fermentation of the hemp, which brought about a slight increase in the cellulose content (see Table 1).



Figure 1. Width and quantitative distribution of the hemp fibres, using FiberShape.

The graphic positioning of the values in the CIELab1976 colour space [109] is presented to enable the best possible differentiation of the differences in chromaticity and luminosity as it is showed in [53,110]. In this research study, colorimetric analysis is of decisive importance to help understand the variations in the chromaticity and the luminosity of the freshly harvested hemp samples, as compared to those preserved in damp anaerobic conditions, which undergo a generalized darkening as a consequence of their storage conditions. Significant variations can also be observed from the yellowish (freshly harvested hemp) to the greenish colour tones (anaerobic wet preserved). This should be taken into consideration in the production of hemp-based products (see Figure 2).



Figure 2. Visual comparison of the two types of hemp (freshly harvested and wet-anaerobic), and their colour representation according to CIELab1976 scheme [109].

The inorganic components used in the samples were studied using X-Ray Diffraction (XRD) and X-Ray Fluorescence (XRF). Their chemical and mineralogical composition can be seen in Table 2 and Figure 3. These are Glass aggregate, which was provided by a Spanish company called Silmin Ibérica (Rubí, Spain). The glass used as an aggregate in the mixture came in the form of a white glass powder with a particle size of 80 to 100 microns. The clay came from a quarry in Guadix (Granada, Spain). Sodium hydroxide (NaOH, in a 5% solution) was supplied by Kremer Pigmente (Aichstetten, Germany) and the liquid sodium silicate (Na₂SiO₃) was supplied by QuiMmipur S.L.U. (Madrid, Spain). For the XRD testing, the raw materials were analysed using a Bruker D8 DISCOVER diffractometer

featuring a DECTRIS PILATUS3R 100K-A detector, from the Granada University Scientific Instruments Centre (CIC, Granada, Spain). The Xpowder program [111] was used to determine its composition. For its part, the X-ray fluorescence (XRF) testing was performed using a high-performance compact wavelength dispersive X-ray Fluorescence Spectrometer (brand PANalytical, model Zeltium, Granada University Scientific Instruments Centre, Granada, Spain).

Table 2. XRF analysis of raw materials (wt %). Data normalized to 100% (LOI-free).

Samples	SiO_2	Al_2O_3	Fe ₂ O ₃	MnO	MgO	CaO	Na ₂ O	K ₂ O	TiO ₂	P_2O_5	SO_3	LOI
Glass powder	68.41	1.19	0.2	0.010	3.08	10.27	14.86	0.46	0.09	0.02	0.30	0.98
Sodium silicate	48.08	0.19	0.03			0.03	19.58	0.07	0.01	0.01	0.05	31.78
Sodium silicate + glass powder	50.41	0.33	0.05		0.51	1.31	18.27	0.14	0.02	0.01	0.08	28.73
Clay	53.80	24.52	7.89	0.06	0.92	1.20	1.32	3.42	0.86	0.22	0.09	5.48



Figure 3. X-ray diffractogram for the clay. Legend: Qz = quartz; Pg = paragonite; Srp = serpentine; Ms = muscovite.

A total of 72 samples was prepared with different formulations. The names and the doses of the groups of test samples are indicated in Table 3. The formulations were given the following abbreviated names: SH1, (Freshly harvested hemp, geopolymer concrete dosage 1); SA2, (anaerobic wet-preserved hemp, geopolymer concrete dosage 2); SH3, (Freshly harvested hemp, geopolymer concrete dosage 3); and SA4, (anaerobic wet-preserved hemp, geopolymer concrete dosage 4). On this basis, all the participating materials without adhesive capacity were considered as aggregates. The amount of additional water used in the SH1, SA2, SH3, and SA4 mixes has been considered in terms of the corresponding percentage of NaOH added. In order to enable us to use them in our experiments, the freshly harvested hemp and the anaerobic wet-preserved hemp were thawed out at T = 22 °C in closed plastic containers 12 h before preparing the concrete for the samples. Prior to mixing, all the materials were stored under the following conditions: T = 22 °C and HR = 70%.

Table 3. Composition and dosage of the samples of geopolymer hempcrete (% volume).

Sample	Freshly Harvested Hemp	Anaerobic Wet-preserved Hemp	Clay (≤1 mm)	Na ₂ SiO ₃	Glass Powder	NaOH
SH1	62.00		21.00	12.00	3.75	1.25
SA2		62.00	21.00	12.00	3.75	1.25
SH3	66.00		10.5	16.00	5.00	2.50
SA4		66.00	10.5	16.00	5.00	2.50

In order to prepare the formulation for the geopolymer hempcrete, we began by preparing the mix of liquid components, to which the sieved clay and the hemp would later be added. To this end, the sodium silicate was mixed with the sodium hydroxide (5% solution in water). The glass dust was then added to this mixture, which was then mixed manually. Once the geopolymer had been homogeneously mixed, the previously sieved (1 mm) clay was added, obtaining a thick suspension to which the hemp was added last of all. The final mixture was then mixed for 3 to 5 min. Two versions were made of each of the three different formulations: one with fresh hemp and the other with 6-month wet-preserved hemp. After removing the moulds, the samples of geopolymer hempcrete were dried on a grid for 12 h at T = 22 °C and HR = 70%. In line with [29,112,113] and previous research [110,114], they were then cured in a drying oven for 24 h at T = 80 °C. After removal from the oven, the samples were placed on a grid where the drying process then continued in the following conditions: T = 22 °C and HR = 70% (see Figure 4).



80 °C (T) - 15% (RH) / 24 h



Geopolymer demoulding and drying

Geopolymer oven hardening

Figure 4. Demoulding, drying, and curing of the geopolymer concrete samples in an oven.

The mineralogy, texture and microstructure of the samples was examined with a scanning electron microscope (SEM) (MODEL Zeiss DMS 950 coupled with Microanalysis Link QX 2000, ZEISS, Jena, Germany) and a polarized optical microscope (MODEL Olympus BX-60, Waltham, MA, USA).

After 28 days, the mechanical resistance of the samples was measured at ambient temperature. Resistance to compression was measured in three 40-mm cube samples, while $40 \times 40 \times 160$ mm prisms were used to measure resistance to bending. These are the standard sizes [115] used in resistance tests. The test was carried out in a universal INSTRON tester (Model 3365) (Instron, Norwood, MA, USA). All the dimensions of each sample were measured (height, length, width and weight), and later, the bulk density was calculated according to standard [116]. The conditions to which the samples were exposed were: temperature = 20 °C and relative humidity = 60%. Finally, the pore structure of the samples was assessed using Mercury Intrusion Porosimetry (MIP).

3. Results

3.1. SEM

The microphotographs shown in the morphological analysis (see Figure 5) reveal several certain differences between the two groups of hemp samples, and in particular the thickening of the hemp fibres.

The top left and right images show the SH1 and SA2 geopolymer concrete, respectively. Swelling of the hemp fibres is indicated with white arrows. The pores are visibly connected, with sizes from 100 μ m. The EDX analyses confirm the good composition of the SH1 and SA2 geopolymer concretes. The comparison between the geopolymer made with freshly harvested hemp (SH3) and the one made with anaerobic wet-preserved hemp (SA4) shows a thickening of the hemp fibres in the latter. Finally, image SA2 shows the polymerization typical of materials of this kind due to the composition and the curing conditions (see white arrows). In addition, lumps of silica can be observed in SA4 (white arrows).



Figure 5. Scanning electron microscopy (SEM) images and EDX analyses.

3.2. Porosimetry

The SH1 and SA2 samples had higher porosity values than samples SH3 and SA4. These higher porosity values are due to the different morphology and distribution of the pores. None of the pores in the samples are closed and the porosity is open. In Figure 6, the trimodal curves show the pore size classification. These curves include very high percentage values (from 81.5% to 91%) for the following pore radius ranges: 100–10 μ m, 10–1 μ m, and 1–0.1 μ m. Pores within these size ranges could be defined as mezzopores [117–119]. Two other types of pores can also be observed: macropores 1000–100 μ m, with values between 2.5% and 7%, and micropores, which ranged from 6.5% to 10.5%, for the following size ranges: 0.1–0.01 μ m, and greater than 0.01 μ m.



Figure 6. Porosimetric values of the samples studied. The relative volume of intruded Hg is shown in brown. The blue path represents the accumulated volume of intruded Hg (Pt = total porosity).

The percentage of pores in the macropore radius range (r) between 1000–100 μ m in anaerobic wet-conserved hemp (A) is half that in freshly harvested hemp (H). In all the

samples, more than 90% of the pores are large or medium-sized. The total porosity values are higher, although the freshly harvested hemp (H) has lower values than the anaerobic wet-preserved hemp (A).

3.3. Density

The density of the geopolymer hempcretes varies between 1061 (\pm 5.75) kg/m³ and 1112 (\pm 10.20) kg/m³ for formulations SH1 and SA2 and between 905 (\pm 20.85) kg/m³ and 912 (\pm 32.12) kg/m³ for formulations SH3 and SA4. These results are shown in Table 4. The results for freshly harvested hemp indicate that these formulations have lower density values than the anaerobic wet-preserved hemp. This is due to the different clay dosage, as higher ratios were used for the SH1-SA2 sample groups than for the SH3-SA4 sample groups.

Table 4. Density and Mechanical Test results.

Samples	Density (kg/m ³)		Flexural Str	rength (MPa)	Compressive Strength (Mpa)		
	$\overline{\mathbf{x}}$	σ	$\overline{\mathbf{x}}$	σ	$\overline{\mathbf{x}}$	σ	
SH1	1061.35	5.75	2.43	0.42	2.14	0.25	
SA2	1112.56	10.20	3.50	0.24	2.57	0.24	
SH3	905.57	20.85	2.20	0.22	1.93	0.02	
SA4	912.36	32.12	3.74	0.60	2.67	0.12	

 \overline{X} : mean values; σ : standard deviation.

3.4. Mechanical Tests

In the compressive strength tests, the SH1 and SA2 formulations obtained values of 2.14 and 2.57 MPa, while SH3 and SA4 obtained scores of 1.93 and 2.67 MPa, respectively. In flexural strength, SH1 and SA2 obtained values of 2.43 and 3.50 MPa, while for SH3 and SA4, these results were 2.20 and 3.74 MPa, respectively (see Table 4).

It must be emphasized that in all the geopolymer concretes, the results in both resistance tests were lower in freshly harvested hemp (H) than in hemp that was wet-preserved in anaerobic conditions for 6 months. The percentage differences are highlighted. As for the formulations of SH1 and SH3, in terms of compression resistance, the H samples obtained values that were 17% and 28% lower than the A samples. Similar differences of 32% and 42% were obtained in bending resistance. In general, the results indicate that the SH1 and SH3 samples achieve lower mechanical results in resistance to bending and to compression than the SA2 and SA4 samples (See Figure 7).



Figure 7. Results of the compressive and bending strength tests.

4. Discussion

When comparing the performance of freshly harvested hemp (H) with that of anaerobic wet-conserved samples (A), the fact that the latter has a lower percentage of cellulose (4%) is the most important reason explaining why the A samples obtained better scores in the mechanical resistance tests.

In this case, their properties improved because of the new structural disposition of the fibres. The results show important differences between the H and A samples of almost 10% in both types of resistance. The research carried out in [82] confirms that the increase in resistance to uniaxial compression and to bending in the anaerobic wet-preserved hemp (A) is due to the existence of cellulose nanocrystals. Along similar lines, [38] indicated that the new conditions of the fibres and the material (a more flexible, lighter, harder biomaterial with very high elasticity modulus and rigidity values) are caused by the increase in the volume of cellulose nanocrystals (CNCs) in the cell walls of certain plant species. Other changes in the fibres can be observed using FiberShape scanning. These include the thickening of the fibres in the anaerobic wet-preserved hemp (after 6 months) compared with the freshly harvested hemp. Moreover, in line with [38,120], the anaerobic wet-preserved hemp (A) is easier to work than the freshly harvested hemp because of the increase in the percentage of cellulose that takes place during the storage process. The changes in the content of hemicellulose and lignin improve the rheology behaviour of the mixtures. The most important changes can be identified by comparing them with the changes that take place when pre-treatments [33] to improve the performance and the properties of the fibres are applied. The results of this study show various benefits in samples conserved in wet anaerobic conditions (A) as compared to the samples of freshly harvested hemp (H).

The relationship between the density and porosity values of the sample groups is dependent on the different proportions of clay used in the formulation of the mixtures. It is also important to remember that there is significant heterogeneity in all the sample groups due to the significant heterogeneity of the hemp and its pore variability.

In general, although the differences between the groups of samples in terms of density and mechanical values are small, the groups of samples with a higher percentage of clay in their dosages have higher densities, as a result of the higher density of the silicates of which they are composed.

This research has also shown that the higher proportion of binder present in SH1 and SA4 formulations causes them to be more resistant to bending than SH3 and SA2, although the difference is small in percentage terms.

It is not clear whether the mechanical strength test values and the density values are related due to the small differences between the different sample groups; what is confirmed is that hemp concrete is highly heterogeneous [47]. However, according to [38,70,82,120–122], the sample groups with anaerobic wet-preserved hemp have better compressive and flexural strength values. Finally, our results show that the doses used in this study led to better mechanical strength results than the dosages used in other similar investigations [123–126].

5. Conclusions

In this paper, we studied the improvement in the behaviour of geopolymers made with wet-preserved hemp in different mixtures. In general, the tests offer positive results and confirm that the hemp conservation method affects the performance of the material. As regards the properties of hemp concrete, we can conclude that the wet anaerobic state of conservation of the hemp improves its qualities as an aggregate, so enhancing the performance of the hempcrete. Specifically, the higher cellulose content in the samples that contained anaerobic wet-preserved hemp improved their mechanical resistance properties. The increases in the percentage of lignin and hemicellulose improved the rheology of the concretes, and by extension, their workability and plasticity. The improvements obtained by anaerobic wet conservation offer important benefits in the use of green concretes of this kind, which in turn offer a range of different benefits in terms of the circular economy. In addition, in this study, it was confirmed that anaerobic wet preservation reduces the volume of water needed to produce this type of green concrete compared to the formulations used by other authors who used almost thirty times more water in the preparation of their samples [58,124,126]. These results were confirmed by the excellent workability of the samples and the small amounts of water required for mixing. This is obviously an important advantage in areas with limited water resources.

Furthermore, this type of conservation means that the fibres can be preserved in suitable conditions for use at any time of year. This could be very useful in countries where it is difficult to grow these plants due to climate conditions. In this way, the maximum potential of the hemp fibres can be secured, because they can be used in good conservation conditions over a much longer period. Finally, this type of preservation has been considered similar to other pre-treatments required to improve the conditions of the hemp fibres prior to use. Therefore, if these pre-treatments are no longer necessary due to the application of this method, the benefits will be even more significant, firstly, by eliminating the consumption of resources required in these pre-treatments, and secondly, by reducing the hemp production costs and by extension those of the final product.

Author Contributions: Conceptualization, M.P.S.-P. and J.A.D.-S.; methodology, M.P.S.-P. and J.A.D.-S.; validation, M.P.S.-P. and J.A.D.-S.; formal analysis, M.P.S.-P. and J.A.D.-S.; investigation, M.P.S.-P. and J.A.D.-S.; resources, M.P.S.-P., J.A.D.-S., and J.C.-G.; data curation, M.P.S.-P. and J.A.D.-S.; writing—original draft preparation, M.P.S.-P. and J.A.D.-S.; writing—review and editing, M.P.S.-P., J.A.D.-S. and J.C.-G.; supervision, M.P.S.-P., J.A.D.-S. and J.C.-G. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Data Availability Statement: Not applicable.

Acknowledgments: This work was supported by the REMINE Project Programme for Research and Innovation Horizon 2020 Marie Skłodowska-Curie Actions, Horizon 2020, WARMEST Project Research and Innovation Staff Exchange (RISE) H2020-MSCA-RISE-2017, RRRMaker project Marie Skłodowska-Curie Research and Innovation Staff Exchange and Project MAT2016-75889-R of the National Plan for Scientific Research, Development, Technological Innovation (Ministry of Science and Technology) and Scientific Unit of excellence "Ciencia en la Alhambra", ref. UCE-PP2018-01, University of Granada and was carried out under the auspices of Research Groups RNM 0179, HUM 629 of the Junta de Andalucía.

Conflicts of Interest: The authors declare no conflict of interest.

References

- 1. Schiavoni, S.; D'Alessandro, F.; Bianchi, F.; Asdrubali, F. Insulation materials for the building sector: A review and comparative analysis. *Renew. Sustain. Energy Rev.* 2016, *62*, 988–1011. [CrossRef]
- Zhang, Y.; Wang, J.; Hu, F.; Yuanfeng, W. Comparison of evaluation standards for green building in China, Britain, United States. *Renew. Sustain. Energy Rev.* 2017, 68, 262–271. [CrossRef]
- Galzerano, B.; Formisano, A.; Durante, M.; Lucolano, F.; Caputo, D.; Liguori, B. Hemp reinforcement in lightweight geopolymers. J. Compos. Mater. 2018, 52, 2313–2320. [CrossRef]
- 4. Alomayri, T.; Shaikh, F.; Low, I.M. Thermal and mechanical properties of cotton fabric-reinforced geopolymer composites. *J. Mater. Sci.* 2013, *48*, 6746–6752. [CrossRef]
- Korniejenko, K.; Frączek, E.; Pytlak, E.; Adamski, M. Mechanical Properties of Geopolymer Composites Reinforced with Natural Fibers. *Procedia Eng.* 2016, 151, 388–393. [CrossRef]
- Assaedi, H.; Shaikh, F.; Low, I.M. Effect of nanoclay on durability and mechanical properties of flax fabric reinforced geopolymer composites. J. Asian Ceram. Soc. 2017, 5, 62–70. [CrossRef]
- Korniejenko, K.; Łach, M.; Hebdowska-Krupa, M.; Mikuła, J. The mechanical properties of flax and hemp fibres reinforced geopolymer composites. *IOP Conf. Ser.-Mater. Sci. Eng.* 2018, 379, 012023. [CrossRef]
- Korniejenko, K.; Łach, M.; Mikuła, J. Mechanical Properties of Raffia Fibres Reinforced Geopolymer Composite. In Advances in "Natural Fibre Composites"; Fangueiro, R., Rana, S., Eds.; Springer: Cham, Switzerland, 2018.
- He, Z.; Zhu, X.; Mu, M.; Wang, Y. Comparison of CO₂ emissions from OPC and recycled cement production. *Constr. Build. Mater.* 2019, 211, 965–973. [CrossRef]

- Huiskes, D.M.A.; Keulen, A.; Yu, Q.L.; Brouwers, H.J.H. Design and performance evaluation of ultra-lightweight geopolymer concrete. *Mater. Des.* 2016, 89, 516–526. [CrossRef]
- Bernhardt, D.; Reilly, J.F. *Mineral Commodity Summaries*; U.S. Geological Survey U.S. Department of the Interior: Washington, DC, USA, 2019; Volume 3, ISBN 9781411342835.
- 12. Amin, M.N.; Ahmad, W.; Khan, K.; Ahmad, A. A Comprehensive Review of Types, Properties, Treatment Methods and Application of Plant Fibers in Construction and Building Materials. *Materials* **2022**, *15*, 4362. [CrossRef]
- Jami, T.; Karade, S.R.; Singh, L.P. A review of the properties of hemp concrete for green building applications. J. Clean. Prod. 2019, 239, 117852. [CrossRef]
- 14. Zuabi, W.; Memari, A.M. Review of Hempcrete as a Sustainable Building Material. *Int. J. Archit. Eng. Constr.* **2021**, *10*, 1–17. [CrossRef]
- 15. Walker, R. A Study of the Properties of the Lime-Hemp Concrete with Pozzolans; Trinity College: Dublin, Ireland, 2013.
- Jami, T.; Kumar, S. Assessment of Carbon Sequestration of Hemp Concrete. In Proceedings of the International Conference on Advances in Construction Materials and Systems, Chennai, India, 3–8 September 2017. [CrossRef]
- 17. Di Capua, S.E.; Paolotti, L.; Moretti, E.; Rocchi, L.; Boggia, A. Evaluation of the Environmental Sustainability of Hemp as a Building Material, through Life Cycle Assessment. *Environ. Clim. Technol.* **2021**, *25*, 1215–1228. [CrossRef]
- 18. Williams, J.; Lawrence, M.; Walker, P. The influence of the casting process on the internal structure and physical properties of hemp-lime. *Mater. Struct.* 2017, *50*, 108. [CrossRef]
- 19. Barbhuiya, S.; Das, B.B. A comprehensive review on the use of hemp in concrete. *Constr. Build. Mater.* **2022**, 341, 127857. [CrossRef]
- 20. Nayana Manohari, T.; Sunil, H.; Rani, D.; Kumar, A. Manufacturing of building blocks using Hempcrete. *Int. J. Latest Res. Eng. Technol.* 2016, 2, 62–73.
- 21. Ruus, A.; Koosapoeg, T.; Pau, M.; Kalamees, T.; Põldaru, M. Influence of production on hemp concrete hygrothermal properties: Sorption, water vapour permeability and water absorption. *J. Phys.: Conf. Ser.* **2021**, 2069, 012004. [CrossRef]
- Fernea, R.; Manea, D.L.; Plesa, L.; Iernuțan, R.; Dumitran, M. Acoustic and thermal properties of hemp-cement building materials. Procedia Manuf. 2019, 32, 208–215. [CrossRef]
- Bennai, F.; Issaadi, N.; Abahri, K.; Belarbi, R.; Tahakourt, A. Experimental characterization of thermal and hygric properties of hemp concrete with consideration of the material age evolution. *Heat Mass Transf.* 2017, 54, 1189–1197. [CrossRef]
- Collet, C.; Bart, M.; Serres, L.; Miriel, J. Porous structure and water vapour sorption of hemp-based materials. *Constr. Build. Mater.* 2008, 22, 1271–1280. [CrossRef]
- 25. Brzyski, P.; Barnat-Hunek, D.; Suchorab, Z.; Lagod, G. Composite materials based on hemp and flax for low-energy buildings. *Materials* 2017, *10*, 510. [CrossRef] [PubMed]
- 26. Latif, E.; Lawrence, M.; Shea, A.; Walker, P. Moisture buffer potential of experimental wall assemblies incorporating formulated hemp-lime. *Build. Environ.* 2015, *93*, 199–209. [CrossRef]
- 27. Dhakal, U.; Berardi, U.; Gorgolewski, M.; Richman, R. Hygrothermal performance of hempcrete for Ontario (Canada) buildings. *J. Clean. Prod.* **2017**, 142, 3655–3664. [CrossRef]
- Colinart, T.; Lelievre, D.; Glouannec, P. Experimental and numerical analysis of the transient hygrothermal behavior of multilayered hemp concrete wall. *Energy Build.* 2015, 112, 1–11. [CrossRef]
- 29. Mustafa Al Bakri, A.M.; Kamarudin, H.; BinHussain, M.; Khairul Nizar, I.; Zarina, Y.; Rafiza, A.R. The Effect of Curing Temperature on Physical and Chemical Properties of Geopolymers. *Phys. Procedia* **2011**, *22*, 286–291. [CrossRef]
- Viel, M.; Collet, F.; Lecieux, Y.; François, M.L.M.; Colson, V.; Lanos, C.; Hussain, A.; Lawrence, M. Resistance to mold development assessment of bio-based building materials. *Compos. Part B Eng.* 2018, 158, 406–418. [CrossRef]
- 31. Duxson, P.; Fernandez-Jimenez, A.; Provis, J.L.; Lukey, G.C.; Palomo, A.; Van Deventer, J.S.J. Geopolymer technology: The current state of the art. *J. Mater. Sci.* 2007, 42, 2917–2933. [CrossRef]
- 32. Provis, J.L.; Bernal, S.A. Alkali Activated Materials: State of the Art Report; Taylor Francis: Abingdon-on-Thames, UK, 2014.
- Malenab, R.A.J.; Ngo, J.P.S.; Promentilla, M.A.B. Chemical Treatment of Waste Abaca for Natural Fiber-Reinforced Geopolymer Composite. *Materials* 2017, 10, 579. [CrossRef]
- 34. Pacheco-Torgal, F.; Castro-Gomes, J.; Jalalic, S. Alkali-activated binders: A review: Part 1. Historical background, terminology, reaction mechanisms and hydration products. *Constr. Build. Mater.* **2008**, *22*, 1305–1314. [CrossRef]
- 35. Pacheco-Torgal, F.; Castro-Gomes, J.; Jalalic, S. Alkali-activated binders: A review. Part 2. About materials and binders manufacture. *Constr. Build. Mater.* 2008, 22, 1315–1322. [CrossRef]
- 36. Alzeer, M.; MacKenzie, K. Synthesis and mechanical properties of novel composites of inorganic polymers (geopolymers) with unidirectional natural flax fibres (phormium tenax). *Appl. Clay Sci.* **2013**, 75–76. [CrossRef]
- 37. Naidu, A.L.; Jagadeesh, V.; Bahubalendruni, M.V.A.R. A review on chemical and physical properties of natural fiber reinforced composites. *Int. J. Adv. Res. Eng. Technol.* **2017**, *8*, 56–68.
- Walbrück, K.; Maeting, F.; Witzleben, S.; Stephan, D. Natural Fiber-Stabilized Geopolymer Foams—A Review. *Materials* 2020, 13, 3198. [CrossRef]
- 39. Bribián, I.Z.; Capilla, A.V.; Usón, A.A. Life cycle assessment of building materials: Comparative analysis of energy and environmental impacts and evaluation of the ecoefficiency improvement potential. *Build. Environ.* **2011**, *46*, 1133–1140. [CrossRef]

- 40. Murri, A.N.; Medri, V.; Landi, E. Production and thermomechanical characterization of wool-geopolymer composites. *J. Am. Ceram. Soc.* **2017**, *100*, 2822–2831. [CrossRef]
- 41. Mishra, J.; Panigrahib, R. Mini-Review on Structural Performance of Fiber Reinforced Geopolymer Concrete. *Int. J. Innov. Technol. Interdiscip. Sci.* **2020**, *3*, 435–442. [CrossRef]
- 42. Verdolotti, L.; Liguori, B.; Capasso, I.; Domenico, E.; Marino, C.; Lavorgna, S. Synergistic effect of vegetable protein and silicon addition on geopolymeric foams properties. J. Mater. Sci. 2015, 50, 2459–2466. [CrossRef]
- 43. Liguori, B.; Capasso, I.; Romeo, V.; D'Auria, M.; Lavorgna, M.; Caputo, D.; Iannace, S.; Verdolotti, L. Hybrid geopolymeric foams with diatomite addition: Effect on chemicophysical properties. *J. Cell. Plast.* **2017**, *53*, 525–536. [CrossRef]
- 44. Nath, P.; Sarker, P.K. Effect of GGBFS on setting, workability and early strength properties of fly ash geopolymer concrete cured in ambient condition. *Constr. Build. Mater.* **2014**, *66*, 163–171. [CrossRef]
- 45. Lee, N.K.; Lee, H.K. Setting and mechanical properties of alkali-activated fly ash/slag concrete manufactured at room temperature. *Constr. Build. Mater.* **2013**, *47*, 1201–1209. [CrossRef]
- Benmahiddine, F.; Bennai, F.; Cherif, R.; Belarbi, R.; Tahakourt, A.; Abahri, K. Experimental investigation on the influence of immersion/drying cycles on the hygrothermal and mechanical properties of hemp concrete. *J. Build. Eng.* 2020, 32, 101758. [CrossRef]
- 47. Belarbi, R.; Bennai, F.; Ferroukhi, M.; Hachem, C.; Abahri, K. Multiscale modelling for better hygrothermal prediction of porous building materials. *MATEC Web Conf.* **2018**, *149*, 02005. [CrossRef]
- Abahri, K.; El Hachem, C.; Bennai, F.; Ngoc, T.; Belarbi, R. Prediction of Hemp Concrete Morphological Deformation by X-ray Tomography. Am. Concr. Inst. ACI Spec. Publ. 2017, 320, 616–625.
- 49. Bennai, F.; Abahri, K.; Belarbi, R.; Tahakourt, A. Periodic homogenization for heat, air, and moisture transfer of porous building materials. *Numer. Heat Transf. Part B Fundam.* **2016**, *70*, 420–440. [CrossRef]
- 50. Maichin, P.; Suwan, T.; Jitsangiam, P.; Chindaprasirt, P.; Fan, M. Effect of self treatment process on properties of natural fiber-reinforced geopolymer composites. *Mater. Manuf. Process.* **2020**, *35*, 1120–1128. [CrossRef]
- 51. Brümmer, M.; Sáez-Pérez, M.P.; Durán Suárez, J.A. *Advances in Natural Fibre Composites*; Fangueiro, R., Rana, S., Eds.; Springer International Publishing: Cham, Switzerland, 2018.
- Merta, I.; Poletanovic, B.; Dragas, J.; Carevic, V.; Ignjatovic, I.; Komljenovic, M. The Influence of Accelerated Carbonation on Physical and Mechanical Properties of Hemp-Fibre-Reinforced Alkali-Activated Fly Ash and Fly Ash/Slag Mortars. *Polymers* 2022, 14, 1799. [CrossRef]
- 53. Delannoy, G.; Marceau, S.; Gle, F.; Gourlay, E.; Guéguen-Minerbe, M.; Diafi, D.; Amziane, S.; Farcas, F. Impact of hemp shiv extractives on hydration of Portland cement. *Constr. Build. Mater.* **2020**, 244, 118300. [CrossRef]
- 54. Niyigena, C.; Amziane, S.; Chateauneuf, A. Multicriteria analysis demonstrating the impact of shiv on the properties of hemp concrete. *Constr. Build. Mater.* **2018**, *160*, 211–222. [CrossRef]
- 55. Magniont, C.; Escadeillas, G. Chemical Composition of Bio-aggregates and Their Interactions with Mineral Binders; Springer: Dordrecht, The Netherlands, 2017; pp. 1–37. [CrossRef]
- 56. Delannoy, G.; Marceau, S.; Glé, P.; Gourlay, E.; Guéguen-Minerbe, M.; Diafi, D.; Nour, I.; Amziane, S.; Farcas, F. Influence of binder on the multiscale properties of hemp concretes. *Eur. J. Environ. Civ. Eng.* **2019**, *23*, 609–625. [CrossRef]
- 57. Piot, A.; Béjat, T.; Jay, A.; Bessette, L.; Wurtz, E.; Barnes-Davin, L. Study of a hempcrete wall exposed to outdoor climate: Effects of the coating. *Constr. Build. Mater.* **2017**, *139*, 540–550. [CrossRef]
- 58. Walker, R.; Pavia, S.; Mitchell, R. Mechanical properties and durability of hemplime concretes. *Constr. Build. Mater.* **2014**, 61, 340–348. [CrossRef]
- 59. Pickering, K.L.; Beckermann, G.W.; Alam, S.N.; Foreman, N.J. Optimising industrial hemp fibre for composites. *Compos. Part A-Appl. Sci. Manuf.* **2007**, *38*, 461–468. [CrossRef]
- 60. Shahzad, A. Hemp fiber and its composites—A review. J. Compos. Mater. 2012, 46, 973–986. [CrossRef]
- 61. Sáez-Pérez, M.P.; Brümmer, M.; Durán-Suárez, J.A. A review of the factors affecting the properties and performance of hemp aggregate concretes. *J. Build. Eng.* 2020, *31*, 101323. [CrossRef]
- 62. Nazerian, M.; Gozali, E.; Dahmardeh, M. The Influence of Wood Extractives and Additives on the Hydration Kinetics of Cement Paste and Cement-bonded Particleboard. *J. Appl. Sci.* 2011, *11*, 2186–2192. [CrossRef]
- 63. Wei, Y.M.; Zhou, Y.G.; Tomita, B. Study of hydration behavior of wood cement-based composite II: Effect of chemical additives on the hydration characteristics and strengths of wood-cement composites. *J. Wood Sci.* **2000**, *46*, 444–451. [CrossRef]
- 64. Del Valle-Zermeño, R.; Aubert, J.E.; Laborel-Préneron, A.; Formosa, J.; Chimenos, J.M. Preliminary study of the mechanical and hygrothermal properties of hemp-magnesium phosphate cements. *Constr. Build. Mater.* **2016**, *105*, 62–68. [CrossRef]
- 65. Nozahic, V.; Amziane, S.; Torrent, G.; Saïdi, K.; De Baynast, H. Design of green concrete made of plant-derived aggregates and a pumice–lime binder. *Cem. Concr. Compos.* **2012**, *34*, 231–241. [CrossRef]
- 66. Ratiarisoa, R.V.; Magniont, C.; Ginestet, S.; Oms, C.; Escadeillas, G. Assessment of distilled lavender stalks as bioaggregate for building materials: Hygrothermal properties, mechanical performance and chemical interactions with mineral pozzolanic binder. *Constr. Build. Mater.* **2016**, *124*, 801–815. [CrossRef]
- 67. Sedan, D.; Pagnoux, C.; Smith, A.; Chotard, T. Mechanical properties of hemp fibre reinforced cement: Influence of the fibre/matrix interaction. *J. Eur. Ceram. Soc.* 2008, 28, 183–192. [CrossRef]

- Balčiunas, G.; Pundiene, I.; Lekunaite-Lukošiune, L.; Vejelis, S.; Korjakins, A. Impact of hemp shives aggregate mineralization on physical-mechanical properties and structure of composite with cementitious binding material. *Ind. Crop Prod.* 2015, 77, 724–734. [CrossRef]
- Diquélou, Y.; Gourlay, E.; Arnaud, L.; Kurek, B. Impact of hemp shiv on cement setting and hardening: Influence of the extracted components from the aggregates and study of the interfaces with the inorganic matrix. *Cem. Concr. Compos.* 2015, 55, 112–121. [CrossRef]
- 70. Maichin, P.; Suwan, T.; Jitsangiam, P.; Chindaprasirt, P. Hemp Fiber Reinforced Geopolymer Composites: Effects of NaOH Concentration on Fiber Pre-Treatment Process. *Key Eng. Mater.* **2020**, *841*, 166–170. [CrossRef]
- Uitterhaegen, E.; Labonne, L.; Merah, O.; Talou, T.; Ballas, S.; Véronèse, T.; Evon, P. Impact of thermomechanical fiber pretreatment using twin-screw extrusion on the production and properties of renewable binderless coriander fiberboards. *Int. J. Mol. Sci.* 2017, *18*, 1539. [CrossRef]
- 72. Bleuze, L.; Lashermes, G.; Alavoine, G.; Recous, S.; Chabbert, B. Tracking the dynamics of hemp dew retting under controlled environmental conditions. *Ind. Crop Prod.* **2018**, *123*, 55–63. [CrossRef]
- Lühr, C.; Pecenka, R.; Budde, J.; Hoffmann, T.; Gusovius, H.J. Comparative investigations of fibreboards resulting from selected hemp varieties. *Ind. Crop Prod.* 2018, 118, 81–94. [CrossRef]
- Sisti, L.; Totaro, G.; Vannini, M.; Celli, A. Retting process as a pretreatment of natural fibres for the development of polymer composites. In *Lignocellulosic Composite Materials*; Kalia, S., Ed.; Springer: Cham, Switzerland, 2018; pp. 97–135.
- Kwunjai, S.; Jitsangiam, P.; Suwan, T.; Rinchumphu, D.; Thongchua, H.; Chindaprasirt, P.; Sampattagul, S. Hemp Fiber Reinforced Geopolymer Composites: Effects of NaOH Concentration on Fiber Pre-Treatment Process. *Key Eng. Mater.* 2020, 841, 171–176. [CrossRef]
- 76. Camargo, M.M.; Adefrs Taye, E.; Roether, J.A.; Tilahun Redda, D.; Boccaccini, A.R. A Review on Natural Fiber-Reinforced Geopolymer and Cement-Based Composites. *Materials* **2020**, *13*, 4603. [CrossRef]
- 77. Nozahic, V.; Amziane, S. Influence of sunflower aggregates surface treatments on physical properties and adhesion with a mineral binder. *Compos. Part A Appl. Sci. Manuf.* **2012**, *43*, 1837–1849. [CrossRef]
- Nunes, C.L. Nonwood bio-based materials. In *Performance of Bio-Based Building Materials*; Woodhead Publishing: Sawston, UK, 2017; pp. 97–196. [CrossRef]
- 79. Anish, M.D.; Mittal, V. Polymer composites with functionalized natural fibers. In *Biodegradable and Biocompatible Polymer Composites*; Processing, Properties and Applications Woodhead Publishing Series in Composites Science and Engineering; Woodhead Publishing: Duxford, UK, 2018. [CrossRef]
- 80. Demir, I.; Doğa, C. Physical and Mechanical Properties of Hempcrete. Open Waste Manag. J. 2020, 13, 26–34. [CrossRef]
- 81. Le Moigne, N.; Oever, M.; Budtova, T. A statistical analysis of fibre size and shape distribution after compounding in composites reinforced by natural fibres. *Compos. Part A* **2011**, *42*, 1542–1550. [CrossRef]
- 82. Gusovius, H.J.; Lühr, C.; Homann, T.; Pecenka, R.; Idler, C. An Alternative to Field Retting: Fibrous Materials Based on Wet Preserved Hemp for the Manufacture of Composites. *Agriculture* **2019**, *9*, 140. [CrossRef]
- 83. Bledzki, A.K.; Gassan, J. Composites reinforced with cellulose based fibres. Prog. Polym. Sci. 1999, 24, 221–274. [CrossRef]
- 84. Bismarck, A.; Mishra, S.; Lampke, T. Plant fibers as reinforcement for green composites. In *Natural fibers biopolymers and biocomposites*; Mohanty, A.K., Misra, M., Drzal, L.T., Eds.; Taylor & Francis: Boca Raton, FL, USA, 2005; pp. 37–108.
- Puglia, D.; Terenzi, A.; Barbosa, S.E.; Kenny, J.M. Polypropylene–natural fibre composites. Analysis of fibre structure modification during compounding and its influence on the final properties. *Compos. Interfaces* 2008, 15, 111–129. [CrossRef]
- 86. Oksman, K.; Mathew, A.P.; Långström, R.; Nyström, B.; Joseph, K. The influence of fibre microstructure on fibre breakage and mechanical properties of natural fibre reinforced polypropylene. *Compos. Sci. Technol.* **2009**, *69*, 1847–1853. [CrossRef]
- 87. Sherin, P.; Nathalie, L.; Deepu, G.; Hanna, J.M.; Ange, N.; Sabu, T. Nanocellulose and its derivative materials for energy and environmental applications. *J. Mater. Sci.* 2022, *57*, 6835–6880. [CrossRef]
- 88. Ferreira, F.V.; Pinheiro, I.F.; de Souza, S.F.; Mei, L.H.I.; Lona, L.M.F. Polymer Composites Reinforced with Natural Fibers and Nanocellulose in the Automotive Industry: A Short Review. *J. Compos. Sci.* **2019**, *3*, 51. [CrossRef]
- 89. Seo, Y.-R.; Kim, J.-W.; Hoon, S.; Kim, J.; Chung, J.H.; Lim, K.-T. Cellulose-based Nanocrystals: Sources and Applications via Agricultural Byproducts. *J. Biosyst. Eng.* 2018, 43, 59–71. [CrossRef]
- Eichhorn, S.J.; Etale, A.; Wang, J.; Berglund, L.A.; Li, Y.; Cai, Y.; Chen, C.; Cranston, E.D.; Johns, M.A.; Fang, Z.; et al. Current international research into cellulose as a functional nanomaterial for advanced applications. *J. Mater. Sci.* 2022, 57, 5697–5767. [CrossRef]
- 91. Tang, Y.; Yang, H.; Vignolini, S. Recent Progress in Production Methods for Cellulose Nanocrystals: Leading to More Sustainable Processes. *Adv. Sustain. Syst.* **2022**, *6*, 2100100. [CrossRef]
- 92. Zhang, Y.; Haque, A.N.M.A.; Naebe, M. Lignin–Cellulose Nanocrystals from Hemp Hurd as Light-Coloured Ultraviolet (UV) Functional Filler for Enhanced Performance of Polyvinyl Alcohol Nanocomposite Films. *Nanomaterials* 2021, 11, 3425. [CrossRef]
- 93. Guo, A.; Sun, Z.; Sathitsuksanoh, N.; Feng, H. A Review on the Application of Nanocellulose in Cementitious Materials. *Nanomaterials* **2020**, *10*, 2476. [CrossRef]
- 94. Pecoraro, M.T.; Mellinas, C.; Piccolella, S.; Garrigos, M.C.; Pacifico, S. Hemp Stem Epidermis and Cuticle: From Waste to Starter in Bio-Based Material Development. *Polymers* 2022, *14*, 2816. [CrossRef]

- 95. Rahmawati, C.; Aprilia, S.; Saidi, T.; Aulia, T.B. Current development of geopolymer cement with nanosilica and cellulose nanocrystals. *J. Phys.: Conf. Ser.* 2021, *1783*, 012056. [CrossRef]
- 96. Kassab., Z.; Abdellaoui, Y.; Hamid Salim, M.; Bouhfid, R.; El Kacem Qaiss, A.; El Achaby, M. Micro- and nano-celluloses derived from hemp stalks and their effect as polymer reinforcing materials. *Carbohydr. Polym.* **2020**, 245, 116506. [CrossRef]
- 97. Tahir, T.M.; Ahmed, A.A.; Saiful Azry, S.O.; Ahmed, Z. Retting process of some bast plant fibres and its effect on fibre quality: A review. *BioResources* 2011, *6*, 5260–5281.
- 98. Liu, M.; Fernando, D.; Daniel, G.; Madsen, B.; Meyer, A.S.; Ale, M.T.; Thygesen, A. Effect of harvest time and field retting duration on the chemical composition, morphology and mechanical properties of hemp fibres. *Ind. Crops Prod.* 2015, 69, 29–39. [CrossRef]
- 99. Idler, C.; Ehlert, D.; Ackermann, I.; Kühne, G. Ernte, Konservierung und Erstverarbeitung von Hanf aus Einer Feuchtgutlinie, (Harvesting, Conservation and First Processing of Hemp from a Moist Material Line). In *Harvesting, Conservation and First Processing of Hemp from a Moist Material Line;* ATB-Projektabschlußbericht 2000/3; Institut für Agrartechnik: Potsdam, Germany, 2000.
- Pecenka, R.; Fürll, C.; Radosavljevic, L. Processing of Wet Preserved Hemp to Fibre Boards in a Pilot Plant. In Proceedings of the International Conference on Flax and Other Bast Plants, Saskatoon, SK, Canada, 20–23 July 2008; pp. 75–80, ISBN 978-0-9809664-0-4.
- Kirilovs, E.; Gusovius, H.J.; Kukle, S.; Emsins, J. Performance of Fibreboards made from Wetpreserved Hemp. *Mater. Sci. Mater. Sci. Text. Cloth. Technol.* 2013, *8*, 65–69. [CrossRef]
- 102. De Maeyer, E.A.A.; Huisman, W. New technology to harvest and store fibre hemp for paper pulp. *J. Int. Hemp Assoc.* **1994**, *1*, 38–41.
- 103. De Maeyer, E.A.A.; Huisman, W. *Techniques for Harvesting and Storage of Fibre Hemp*; IMAG-DLO and Wageningen Agricultural University: Wageningen, The Netherlands, 1995; p. 138, (In Dutch with English summary).
- 104. Sáez-Pérez, M.P.; Brümmer, M.; Durán-Suárez, J.A. Effect of the state of conservation of the hemp used in geopolymer and hydraulic lime concretes. *Constr. Build. Mater.* **2021**, *285*, 122853. [CrossRef]
- Tutek, K.; Masek, A. Hemp and Its Derivatives as a Universal Industrial Raw Material (with Particular Emphasis on the Polymer Industry)—A Review. *Materials* 2022, 15, 2565. [CrossRef] [PubMed]
- 106. Clarence, P.; Ginga, C.P.; Ongpeng, J.M.C.; Daly, M.A.; Klarissa, M. Circular Economy on Construction and Demolition Waste: A Literature Review on Material Recovery and Production. *Materials* **2020**, *13*, 2970. [CrossRef]
- 107. Leibniz Institute for Agricultural Engineering and Bioeconomy (ATB). Available online: https://www.leibniz-gemeinschaft.de/ en/institutes/leibniz-institutes-all-lists/leibniz-institute-for-agricultural-engineering-and-bioeconomy.html (accessed on 29 August 2022).
- 108. Müssig, J.; Schmid, H.G. Quality control of fibres along the value added chain by using scanning technique—From fibres to the final product. *Microsc. Microanal.* 2004, 10, 1332–1333. [CrossRef]
- 109. ISO 11664-4:2008(E)/CIE S 014-4/E:2007; Colorimetry—Part 4: CIE 1976 L*a*b* Colour Space. ISO: Geneva, Switzerland, 2008.
- Sáez-Pérez, M.P.; Durán-Suárez, J.A.; Brummer, M. Hemp concrete for the sustainable retrofit of the vernacular architectural heritage in the region of Senhaja Srair (Morocco). In *Conserving Cultural Heritage*; Taylor and Francis Group: London, UK, 2018.
 Martin, J.D. XPowderX, Qualitative, Quantitative and Microtexture. *XPowderX* 16.01.10 version, 2016.
- 112. Ekaputri, J.J.; Priyanka, N.F. The Effect of Temperature Curing on Geopolymer Concrete. MATEC Web Conf. 2017, 97, 01005.
- [CrossRef] 113. Yang, H.; Zhu, M.C.; Fang, C.Q. Review of research on high-temperature behavior of geopolymer paste. *E3S Web Conf.* **2021**,
- 237, 01017. [CrossRef]
- 114. Beghoura, I.; Castro-Gomes, J. Development of Porous Tungsten Mud Waste-based Alkali-activated Foams with Low Thermal Conductivity. *Constr. Build. Mater.* **2019**, 224, 682–690. [CrossRef]
- 115. *115.* UNE-EN 1015-11:2000/A1:2007; Methods of Test for Mortar for Masonry—Part 11: Determination of Flexural and Compressive Strength of Hardened Mortar. UNE-EN: Madrid, Spain, 2007.
- 116. UNE-EN 12390-7:2020; Testing Hardened Concrete Part 7: Density of Hardened Concrete. UNE-EN: Madrid, Spain, 2020.
- 117. Veniale, F.; Zezza, U. Nuove indagini sull arenaria della Basilicata di S. Michelle in Pavia. *Atti Ticinensi De Sci. Della Terra* **1988**, 31, 253–268.
- 118. Zehnder, K. Verwitterung von Molassesandsteinen an Bauwerken und in Naturaufschlüssen. 130 S. Doctoral Thesis, 78 Textfig. ETH Zürich, Zürich, Switzerland, 1982. [CrossRef]
- 119. Russel, S.A. Stone Preservation Committee Report (Appendix I); H.M. Stationary Office: London, UK, 1927.
- 120. Dri, F.L.; Hector, L., Jr.; Moon, R.J.; Zavattieri, P.D. Anisotropy of the elastic properties of crystalline cellulose Iβ from first principles density functional theory with Van der Waals interactions. *Cellulose* **2013**, *20*, 2703–2718. [CrossRef]
- Lawrence, M.; Heath, A.; Walker, P. Development of a novel mortar for use with unfired clay bricks. *Proc. Inst. Civ. Eng.*—Constr. Mater. 2013, 166, 18–26. [CrossRef]
- 122. Castillo-Lara, J.F.; Flores-Johnson, E.A.; Valadez-Gonzalez, A.; Herrera-Franco, P.J.; Carrillo, J.G.; Gonzalez-Chi, P.I.; Li, Q.M. Mechanical properties of natural fiber reinforced foamed concrete. *Materials* **2020**, *13*, 3060. [CrossRef]
- 123. Fernea, R.; Manea, D.L.; Tamas Gravea, D.R.; Rosca, J.C. Hemp-clay building materials. An investigation on acoustic, thermal and mechanical properties. *Procedia Manuf.* **2019**, *32*, 216–223. [CrossRef]
- 124. Mazhoud, B.; Collet, F.; Pretot, S.; Lanos, C. Mechanical properties of hemp-clay and hemp stabilized clay composites. *Constr. Build. Mater.* **2017**, *155*, 1126–1137. [CrossRef]

- 125. Brümmer, M.; Sáez-Pérez, M.P.; Durán Suárez, J.A. Hemp-Clay Concretes for Environmental Building—Features that Attribute to Drying, Stabilization with Lime, Water Uptake and Mechanical Strength. In *Advances in Natural Fibre Composites*; Fangueiro, R., Rana, S., Eds.; Springer International Publishing: Cham, Switzerland, 2018; pp. 249–265. [CrossRef]
- 126. Balčiūnas, G.; Vėjelis, S.; Vaitkus, S.; Kairytė, A. Physical properties and structure of composite made by using hemp hurds and different binding materials. *Procedia Eng.* **2013**, *57*, 159–166. [CrossRef]