

Article

A Cellulose Aerogel Made from Paper and Hemp Waste Added with Starch for the Sorption of Oil

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Abstract: One sustainable method of removing oil spills is the use of sorbents. In this study, aerogels made from paper waste and crosslinking starch are investigated as sorbents. Aerogels have a high porosity and, thus, achieve a high sorption capacity. The paper waste aerogels are compared with aerogel samples that also contain hemp fibres as a composite. After hydrophobisation with methyltrimethoxysilane, the maximum sorption capacity of crude oil, marine diesel oil and lubricating oil is investigated. Aerogels made of cellulose from paper waste with starch show the highest sorption capacity of about 50 gg^{-1} for all studied oils. Unfortunately, hemp fibres offer no advantage in sorption capacity, but they do mitigate a decrease in sorption capacity with an increase in cellulose fibre content. This could be an advantage in a possible commercial product, so that the properties do not vary as much.

Keywords: cellulose aerogel; starch; hemp fibres; hydrophobisation; sorption capacity; crude oil; marine diesel oil; lubricant oil



Citation: Paulauskiene, T.; Sirtaute, E.; Uebe, J. A Cellulose Aerogel Made from Paper and Hemp Waste Added with Starch for the Sorption of Oil. *J. Mar. Sci. Eng.* **2023**, *11*, 1343. <https://doi.org/10.3390/jmse11071343>

Academic Editor: Theocharis D. Tsoutsos

Received: 7 June 2023

Revised: 26 June 2023

Accepted: 30 June 2023

Published: 30 June 2023



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1. Introduction

Nowadays, human activities contribute significantly to environmental changes. The more advances are made in various technical areas, the more accidents occur. This also includes oil and oil product spills caused by transport and cargo transfers to oil terminals [1].

One of the most sustainable methods of removing oil spills is the use of sorbents [2,3]. The authors investigated hydrophobised cellulose aerogels as a particularly effective sorbent [4–7]. In general, aerogels have a high porosity and, thus, achieve a high sorption capacity [8]. Cellulose-based aerogels are made from a variety of materials, such as corn straw [9], hemp [10], rice husk [11], potato tubers [12], waste paper [13], cotton [14], rice straw [15], timber [16], etc. Plant biomass in forests is the main feedstock for cellulosic aerogels, while renewable resources, such as aquatic plants, grasses and agricultural residues, are considered as an essential source of cellulosic aerogels [17]. Aerogels made from paper waste are particularly sustainable, as the amounts of paper waste are still increasing [18,19]. The paper industry is one of the largest industries in the world [20].

Composites are prepared from at least two constituent materials. In general, fibres in an amorphous matrix are reported, as described, for example, in [21]. A composite of a cellulose aerogel from paper waste with fibres from office paper, cardboard, wool, straw, algae and cellulose acetate is reported in [6]. The wool/cellulose aerogel composite exhibited higher sorption capacities for oil than “pure” cellulose aerogel. Cellulose aerogel with cellulose acetate also showed higher sorption capacities, but it could only be used once. Surprisingly, to the authors’ knowledge, very little literature exists about composites of cellulose aerogels consisting of cellulose fibres and chemically untreated, virgin fibres of hemp straw for oil spill sorbent applications. Hemp straw is an agricultural waste product and is only described in the scientific literature in connection with thermal insulation [22–24], where it is processed into aerogel in a sol-gel process.

In this paper the results of the investigation of aerogels from paper waste and hemp fibres are described, using the samples as sorbents for three kinds of oil after their hydrophobisation. The goal of the incorporation of hemp fibres into cellulosic aerogels is to modify the sorption capacities of cellulosic aerogels by incorporating these materials into the cellulosic fibres for the aerogels, without prior treatment or dissolution. In addition, starch should be used as a sustainable cross-linker [25] which, according to [7], also leads to a significant increase in the sorption capacity of the cellulose aerogel. The aerogel samples are rendered hydrophobic with a silane reagent, creating an oil sorbent, characterised by the contact angle measurements with water drops. This procedure is recommended in the literature [26]. Density and porosity of the produced aerogels were determined. Furthermore, it was examined whether an increase in the sorption properties of crude oil, marine diesel and biodiesel can be measured by the additives. The results of the mechanical property studies will be published elsewhere.

2. Materials and Methods

2.1. Materials

For the synthesis of the aerogels, cardboard waste (4.01.00 code according to EN 643:214 “Paper and board. European list of standard grades of paper and board for recycling”) as the source of cellulose, hemp fibres of approximately 0.5 cm fibre length from a local farmer at Klaipeda (Lithuania), and potato starch (Aloja Starkelsen Ltd., Liepaja, Latvia) were used as raw materials. Methyltrimethoxysilan (MTMS of 98%; Sigma-Aldrich Chemie GmbH, Taufkirchen, Germany) was used for hydrophobisation of aerogels.

For the investigation of the maximum sorption capacities, crude oil with a density of 867 kg m⁻³ and a dynamic viscosity of 0.0097 Pa s from SC ORLEN Lietuva (Mažeikiai, Lithuania), marine diesel oil (MDO) with a density of 852 kg m⁻³ and a dynamic viscosity of 0.0024 Pa s from JSC Gindana (Klaipėda, Lithuania) and the lubricating oil PEMCO iDrive 105 SAE 15 w-40 with a density of 877 kg m⁻³ and a dynamic viscosity of 0.0038 Pa s from JSC SCT Lubricants (Klaipėda, Lithuania) were used.

2.2. Production of the Aerogel Samples

For the control samples, cardboard was shredded up to 0.5 cm² in size, and an amount of 200 mL distilled water (1 wt%) was homogenised in a high beaker at 20,000 rpm for five minutes with a disperser UltraTurrax T25 digital (IKA, stainless steel rotor/stator diameter of 18 mm). In parallel, starch was dissolved in 20 mL hot water. The starch solution was added to the mixture, and homogenised for about one minute. Then, the mixture was poured into a 40 mL metal cups and refrigerated for 3 h at minus 18 °C. The frozen samples were then freeze-dried in a vacuum at 0.015 hPa and a condenser temperature of minus 105 °C (ScanVac CoolSafe; LaboGene, Hadsten, Denmark).

The proportions of starch to cardboard per 100 mL of water can be found in Table 1.

Table 1. Material quantities of starch and cardboard for the control samples per 100 mL batch size.

Starch in (g)	Water in (mL)	Cardboard in (g)	Water in (mL)
0.1		1	
0.2		2	
0.3	20	3	80
0.4		4	
0.5		5	

The procedure of hydrophobisation of aerogel samples follows the one described in [7].

The aerogel samples with the hemp fibres were prepared in the same way as the control samples described above but, instead of a pure starch solution, a mixture of starch and hemp fibres was added as listed in Table 2.

Table 2. Material quantities of starch, hemp fibres, and cardboard for the control samples per 100 mL batch size.

Starch in (g)	Hemp Fibres in (g)	Water in (mL)	Cardboard in (g)	Water in (mL)
0.05	0.05	20	1	80
0.1	0.1	20	2	80
0.15	0.15	20	3	80
0.2	0.2	20	4	80
0.25	0.25	20	5	80

The hydrophobisation of these aerogel samples follows the above-described procedure. A total of 5 samples were produced from each aerogel group.

2.3. Investigation of the Properties the Aerogel Samples

For the density of aerogels, the samples were weighed, and the height and diameter were measured at several surface locations to calculate the density of the aerogels.

The porosity of aerogels is calculated as given in [6]:

$$Porosity = \left(1 - \left(\frac{\rho_s}{\rho_{t.s.}} \right) \right) \cdot 100\%, \tag{1}$$

where ρ_s is the bulk density of the aerogel samples and $\rho_{t.s.}$ is the theoretically density of the mixtures of the starch and the cellulose and hemp fibres, and can be calculated according to [27]:

$$\rho_{t.s.} = \frac{1}{\frac{w_{cellulose}}{\rho_{cellulose}} + \frac{w_{starch}}{\rho_{starch}}} \tag{2}$$

where $w_{cellulose}$ and w_{starch} are the parts of the sum of cellulose, hemp fibres and starch in the sample; and $\rho_{cellulose}$ and ρ_{starch} are the densities of the cellulose and starch, respectively. According to the literature, the density of cellulose is 1.5 g cm^{-3} [28], the density of starch is 1.45 g cm^{-3} [29] and the density of the hemp fibres is assumed to be that of cellulose.

The wettability of the aerogel surface was investigated by measuring the water contact angle using the sessile drop method as described in [7].

The maximum sorption capacity of aerogels (Q) is determined using [6]:

$$Q = \frac{m_1 - m_0}{m_0}, \text{ g g}^{-1} \tag{3}$$

where m_0 is the weight of the aerogel before sorption in g and m_1 is the weight of the aerogel after sorption in g.

3. Results

3.1. Hydrophobicity Test Results of Aerogel Samples

The water contact angle of all samples was determined after hydrophobisation using MTMS. The contact angles of water droplets (Figure 1) were measured at several locations and the results were averaged. It has been observed that all cellulose aerogels have fairly similar water contact angles, mostly around 100° to about 120° (Tables 3 and 4). A dependence on the composition cannot be determined.

Table 3. Water drop angle determined of aerogel samples with different concentrations of waste paper (P) and starch (K).

K0.1-P1	K0.2-P2	K0.3-P3	K0.4-P4	K0.5-P5
$(115 \pm 3)^\circ$	$(122 \pm 4)^\circ$	$(121 \pm 4)^\circ$	$(114 \pm 6)^\circ$	$(109 \pm 5)^\circ$

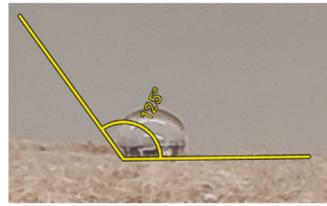


Figure 1. Representative photograph of a water drop on a hydrophobised aerogel made from waste paper and starch.

Table 4. Water droplet angle determined for different concentrations of aerogel samples with waste paper (P), starch (K), and hemp (KA) fibres.

K0.05-KA0.05-P1	K0.1-KA0.1-P2	K0.15-KA0.15-P3	K0.2-KA0.2-P4	K0.25-KA0.25-P5
$(100 \pm 1)^\circ$	$(114 \pm 6)^\circ$	$(114 \pm 6)^\circ$	$(118 \pm 7)^\circ$	$(120 \pm 6)^\circ$

The maximum water contact angle was recorded at 131° at the two cellulose concentrations—K0.3-P3 and K0.4-P4. The smallest water contact angle of 123° was recorded at the sample K0.1-P1.

The maximum water contact angle was recorded at 131° , again at the two cellulose concentrations, namely K0.25-KA0.25-P5, and the smallest, 100° , also in the sample (K0.05-KA0.05-P1).

These contact angles correspond to the values reported in [7]. In general, the contact angles of water to the aerogel samples being higher than 90° means that the samples are hydrophobic [6].

3.2. Determination of Density and Porosity of Aerogel Samples

The density and porosity of control samples from different amounts of cellulose from cardboard waste, and the correlating amount of cross-linker starch, are shown in Figure 2. It can be seen that the lowest density of cellulose-based aerogels using starch reached $15.209 \pm 2.099 \text{ kg m}^{-3}$, where both cellulose and binder concentration in the solution were the lowest (K0.1-P1). On the other hand, the highest value was obtained in sample K0.5-P5, with the highest cellulose and starch content— $80.772 \pm 6.454 \text{ kg m}^{-3}$. The density increases and porosity decreases with increasing content of cellulose. However, porosity does not appear to decrease linearly with increasing cellulose content, just as density inversely increases linearly.

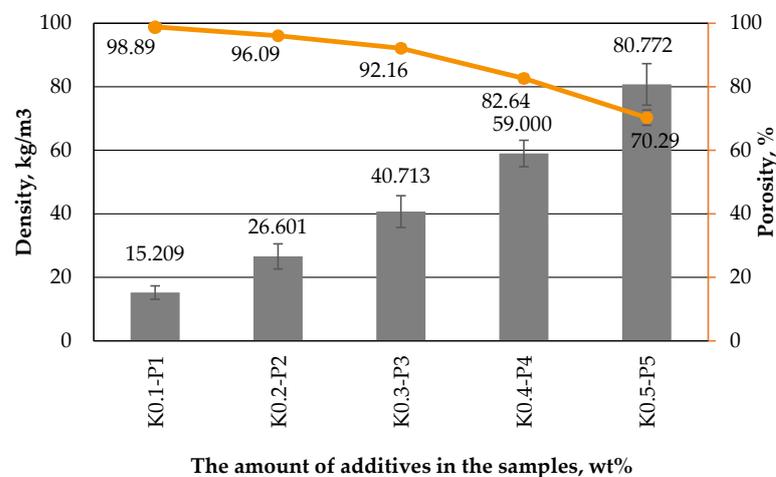


Figure 2. Results of testing the density and porosity of control aerogel samples with starch with different cellulose concentrations: K—starch content in the samples, wt%; P—the amount of paper waste in the samples, wt%.

The non-linear decrease in porosity with increasing cellulose content could be related to the fact that the thickness, as can be seen in the REM images in [7], forms its own network and thus contributes to the porosity.

Figure 3 shows the densities and porosities of the aerogel samples, which also contain hemp fibres. Again, the porosity decreases more slowly than the cellulose content from the waste paper increases.

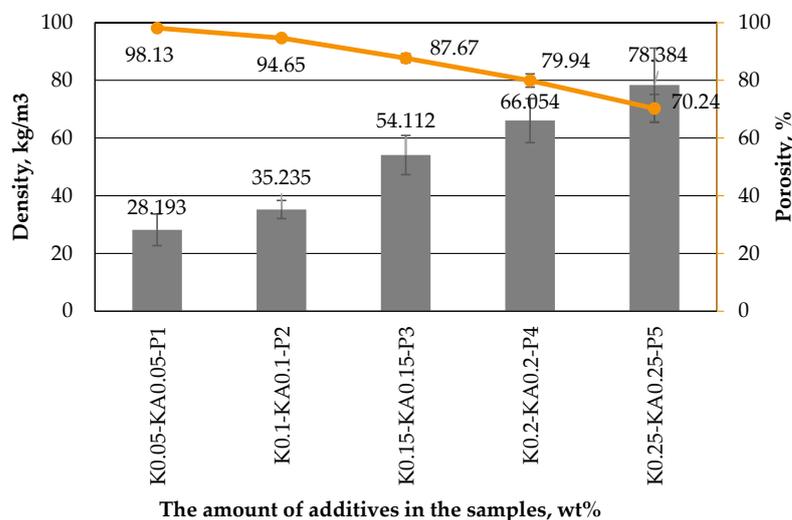


Figure 3. Results of testing the density and porosity of control aerogel samples with starch with different cellulose concentrations: K—starch content in the samples, wt%; KA—the amount of hemp fibre in the samples, wt%; P—the amount of paper waste in the samples, wt%.

The lowest density value was obtained for the sample K0.05-KA0.05-P1 – $28.193 \pm 5.481 \text{ kg m}^{-3}$, and the highest for K0.25-K0.25-P5 – $78.384 \pm 12.793 \text{ kg m}^{-3}$ aerogel samples. However, compared to the control samples from Figure 2, the porosity values here are somewhat lower. This could be explained by the additional hemp fibres. The values of the densities are similar to the range of the values as already determined in [7]. In the literature [26,30–32], a typical aerogel porosity ranges from 80 to 99.8 percent. Porosity varies depending on the amounts of constituents in the sample, their density, and processing methods.

3.3. The Results of the Maximum Sorption Capacity of Aerogels

To determine the sorption capacity of aerogels for oil, 15 samples were prepared for each different cellulose concentration. Five samples of each concentration were used for marine diesel oil, crude oil and lubricating oil tests, respectively. Control samples containing waste paper and starch were tested first, then samples containing additional hemp fibres were tested.

Figure 4 shows the results of the investigation of the maximum sorption capacity of aerogels for the sorption of marine diesel and the influence of the amount and type of the individual additives on the sorption capacity.

As can be seen in Figure 4a, the sorption capacity of the control samples decreases from $49.73 \pm 1.34 \text{ gg}^{-1}$ (control samples) to $10.55 \pm 0.43 \text{ gg}^{-1}$, with increasing concentration of cellulosic additives in the samples from 1 to 5% by weight. Similarly, a trend of decreasing sorption capacity from $34.48 \pm 5.62 \text{ gg}^{-1}$ to $10.76 \pm 1.35 \text{ gg}^{-1}$ is observed in samples with hemp fibre. Since the maximum sorption capacity of oil is related to the pore volume of the aerogels (the larger the pore volume, the higher the maximum sorption capacity [33]), the decreasing trend can be explained by the increased hemp fibre concentration and the consequent lower porosity of the resulting aerogels (see also Figures 2 and 3) and, thus, less space is available for the absorption of marine diesel oil.

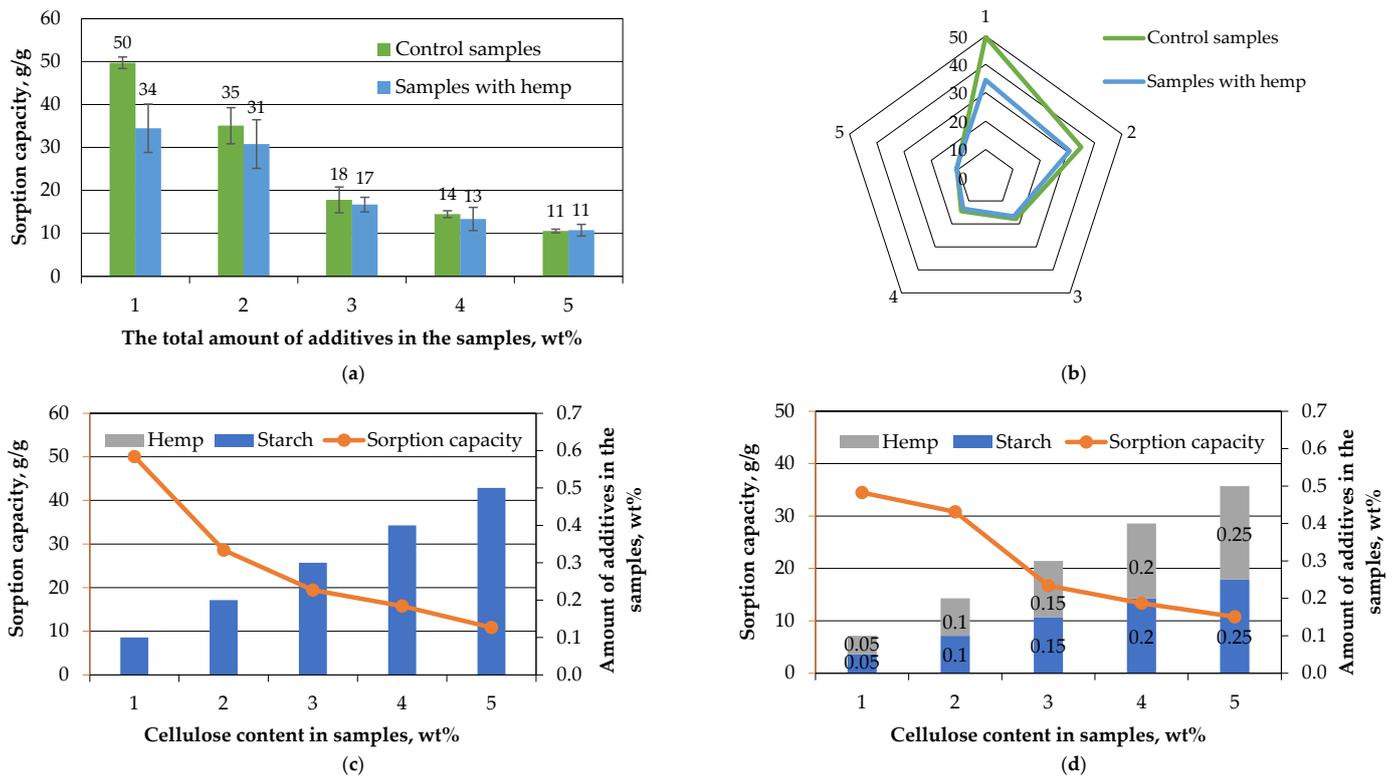


Figure 4. The results of the investigation of the maximum sorption capacity of cellulose aerogel samples for the sorption of marine diesel oil: (a) dependence of sorption capacity on the amounts of additives used for the production of aerogels; (b) comparison of the sorption capacity of control samples and samples with hemp fibre insertion (c,d) the influence of the amounts of individual additives on the sorption capacity of aerogels in control samples and samples with hemp fibre, respectively.

Figure 4 shows the decrease in the maximum sorption capacity of the control samples made from cellulose and starch. The drop of the maximum sorption capacity between 2 and 3 g cellulose content of the control samples is quite high compared to the drops before and after. The samples with hemp fibres differ from the control samples. The maximum sorption capacity of the first sample is similar to the sorption capacity of a sample without additional starch and hemp fibres, as described in [4]. The drop of maximum sorption capacity between the first aerogel samples with 1 and 2 g cellulose content is lower than that of the corresponding control samples. When comparing the maximum sorption capacity of the aerogel samples with hemp fibres with the control samples for marine diesel oil, the difference in the maximum sorption capacity of about 30% is noticeable for the sample with 1 g cellulose content. The difference is significantly smaller for the other samples in Figure 4.

The highest values of the sorption capacity of marine diesel oil were obtained for the control samples and the samples with hemp fibre with a 1–2 wt% concentration of cellulose (see Figure 4b).

In Figure 4c, we see that the maximum sorption capacity of the control sample, which consists of 1 wt% of cellulose and 0.1 wt% of starch, reached $49.73 \pm 1.34 \text{ gg}^{-1}$, while the sorption capacity of the sample, which consisted of 5 wt% of cellulose and 0.5 wt% of starch, reached $10.55 \pm 0.43 \text{ gg}^{-1}$ only. This is almost six times lower compared to the sample with the lowest cellulose concentration.

Comparing the data of Figure 4c,d, the influence of the amounts of individual additives, such as starch and hemp fibres, on the marine diesel oil sorption by aerogels could be seen. The first series of samples consisted of 0.05 wt% of hemp fibre, 0.05 wt% of starch and 1 wt% of cellulose has maximum sorption capacity of $34.48 \pm 5.62 \text{ gg}^{-1}$. Meanwhile, the last batch of samples consisted of 0.25 wt% of hemp fibre, 0.25 wt% of starch and 5 wt%

of cellulose has sorption capacity of $10.76 \pm 1.35 \text{ gg}^{-1}$. Based on the porosity and density research results (see Figures 2 and 3), it can be stated that additionally added hemp fibres increase the density of aerogels and reduces its porosity, which leads to a decrease in the maximum sorption capacity of marine diesel oil.

Figure 5 shows the results of the investigation of the maximum sorption capacity of aerogels for the sorption of crude oil and the influence of the amount and type of the individual additives on the sorption capacity.

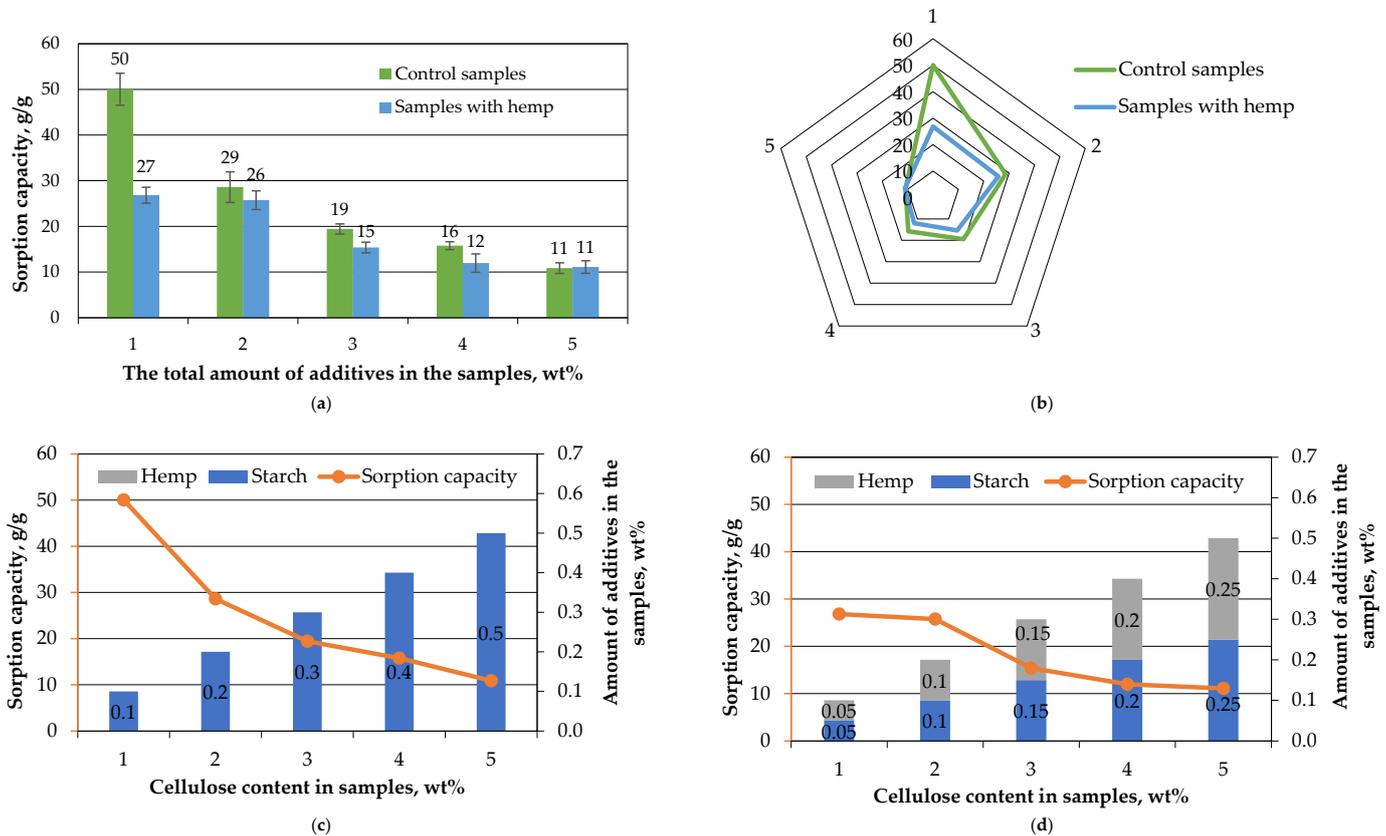


Figure 5. The results of the investigation of the maximum sorption capacity of cellulosic aerogel samples for crude oil sorption: (a) dependence of sorption capacity on the amounts of additives used for the production of aerogels; (b) comparison of the sorption capacity of control samples and samples with hemp fibre insertion (c,d) the influence of the amounts of individual additives on the sorption capacity of aerogels in control samples and samples with hemp fibre, respectively.

Here, too, overall and when comparing the effects of the individual components, a behaviour similar to that of the previously described results on marine diesel oil is revealed. The control sample of 1 g cellulose and 0.1 starch shows a similarly high maximum sorption capacity for crude oil as for marine diesel oil. This result is consistent with the results in [7]. The sorption values also decrease in a similar way with increasing cellulose contents (Figure 5 as in Figure 4). However, the maximum sorption capacity when replacing half of the starch in the aerogel sample with 1 g cellulose decreases more than in the previously described marine diesel oil as sorbate. This could be related to the significantly more complex composition of the crude oil. When comparing the maximum sorption capacity of the aerogel samples with hemp fibres with the control samples for crude oil, the difference in the maximum sorption capacity of about 53% is noticeable for the sample with 1 g cellulose content. The difference is significantly smaller for the other samples, as shown in Figure 5.

The highest values of the sorption capacity of marine diesel oil were obtained in the case of marine diesel oil sorption for control samples, as well as the samples with hemp fibre with a 1 and 2 wt% concentration of cellulose (see Figure 5b).

Comparing the data of Figure 5c,d, it can be seen that the maximum sorption capacity of the first series of samples, consisting of 0.05 wt% of hemp fibre, 0.05 wt% of starch and 1 wt% of cellulose, reached $26.84 \pm 1.73 \text{ gg}^{-1}$, and the last series of samples, consisting of 0.25 wt% of hemp fibre, 0.25 wt% of starch and 5 wt% of cellulose, reached $11.10 \pm 1.37 \text{ gg}^{-1}$ sorption capacity.

The results of the sorption capacity of 1 wt% of cellulose concentration aerogels with the hemp fibre was 22% lower for crude oil sorption ($26.84 \pm 1.73 \text{ gg}^{-1}$), than that of diesel, $-34.48 \pm 5.62 \text{ gg}^{-1}$. Meanwhile, 5 wt% of cellulose and hemp samples crude oil sorption capacity was $11.11 \pm 1.37 \text{ gg}^{-1}$, and was similar to the marine diesel oil sorption capacity results of $10.76 \pm 1.35 \text{ gg}^{-1}$.

Figure 6 shows the results of the investigation of the maximum sorption capacity of aerogels for the sorption of lubricating oil and the influence of the amount and type of the individual additives on the sorption capacity.

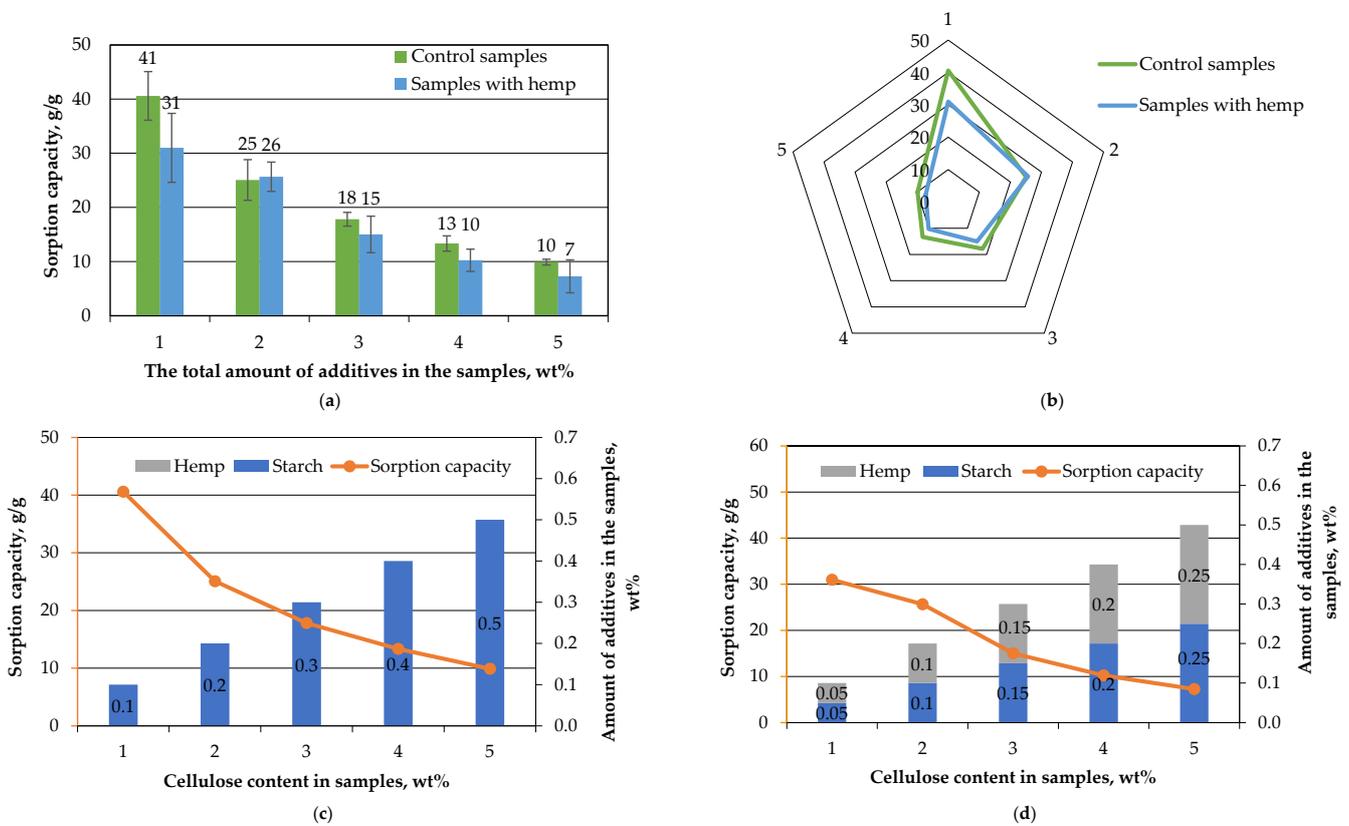


Figure 6. The results of the investigation of the maximum sorption capacity of cellulosic aerogels in the sorption of lubricating oil: (a) dependence of sorption capacity on the amounts of additives used for the production of aerogels; (b) comparison of the sorption capacity of control samples and samples with hemp fibre insertion (c,d) the influence of the amounts of individual additives on the sorption capacity of aerogels in control samples and samples with hemp fibre, respectively.

Here, the control samples in Figure 6 show a more even drop in the maximum sorption capacity for lubricating oil than in the previously described investigations on crude oil and marine diesel oil. However, the maximum sorption capacity for lubricating oil in the control sample with 1 g cellulose and 0.1 g starch is about 20% smaller ($30.99 \pm 6.37 \text{ gg}^{-1}$) than for the other two oils. When comparing the maximum sorption capacity of the aerogel samples with hemp fibres with the control samples for lubricating oil, the difference in the maximum sorption capacity of about 24% is noticeable for the sample with 1 g cellulose content. The difference is significantly smaller for the other samples in Figure 6.

According to the results of studying the sorption capacity of marine diesel oil and crude oil, it can be stated that aerogels with the lowest cellulose concentration mainly absorb marine diesel oil ($34.48 \pm 5.62 \text{ gg}^{-1}$) and 22% less crude oil ($26.83 \pm 1.73 \text{ gg}^{-1}$). Meanwhile, aerogels with the highest concentration of cellulose absorb primarily marine diesel ($10.76 \pm 1.35 \text{ gg}^{-1}$) and nearly 32.5% less lubricating oil ($7.27 \pm 3.05 \text{ gg}^{-1}$). This is also related to the density and viscosity of the sorbed product, and is also confirmed by [4,34].

4. Conclusions

It was successfully demonstrated that composite aerogel samples can be produced from cellulose fibres with untreated hemp fibres and a crosslinking starch. The samples have a low density and a high porosity typical for aerogels.

These samples were then successfully hydrophobised to sorb three different oils. Since starch was introduced as sol into a paper waste slurry, it was able to form its own network, as shown in an earlier publication by the authors. However, the maximum sorption capacities of the composite gels are consistently lower than those for cellulose aerogel with starch and without hemp fibres. This is probably due to the increased density and the therefore reduced porosity caused by the hemp fibres. The maximum sorption capacity of the cellulose aerogel with starch and hemp fibres is similar to that of a cellulose aerogel sample without the addition of starch and hemp fibres. The hemp fibres neutralise the increasing effect of starch, which was found in an early study of the authors. A composite site consisting of cellulose fibres from paper waste and hemp fibres and crosslinking starch, therefore, has no advantage over a “pure” cellulose aerogel with respect to the sorption of oils.

It is nevertheless interesting to note that hemp makes the drop in maximum sorption capacity for aerogels with 1 or 2 g cellulose comparatively small. A small drop in the maximum sorption capacity could be important in a commercial product if there are variations in the weight of the starting materials. Therefore, the authors will search for a substitute for hemp that reduces the drop in sorption capacity similarly, but keeps the absolute sorption capacity for cellulose aerogels at a similar or higher level as for starch aerogel samples.

Author Contributions: Conceptualization, T.P.; methodology, T.P., E.S. and J.U.; software, E.S.; validation, T.P., E.S. and J.U.; formal analysis, E.S.; investigation, E.S.; resources, T.P.; data curation, T.P. and J.U.; writing—original draft preparation, J.U. and E.S.; writing—review and editing, T.P. and J.U.; visualization, T.P. and E.S.; supervision, T.P. and J.U.; project administration, T.P.; funding acquisition, T.P. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: All data generated or analysed during this study are included in this published article.

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

References

1. ITOPF's Annual Tanker Spill Statistics Show a Promising Start to the New Decade. Available online: <https://www.itopf.org/news-events/news/itopfs-annual-tanker-spill-statistics-show-a-promising-start-to-the-new-decade/> (accessed on 4 January 2023).
2. Li, H.; Liu, L.; Yang, F. Oleophilic polyurethane foams for oil spill cleanup. *Procedia Environ. Sci.* **2013**, *18*, 528–533. [CrossRef]
3. Zamparas, M.; Tzivras, D.; Dracopoulos, V.; Ioannides, T. Application of Sorbents for Oil Spill Cleanup Focusing on Natural-Based Modified Materials: A Review. *Molecules* **2020**, *25*, 4522. [CrossRef] [PubMed]
4. Paulauskiene, T.; Uebe, J.; Karasu, A.U.; Anne, O. Investigation of Cellulose-Based Aerogels for Oil Spill Removal. *Water Air Soil Pollut.* **2020**, *231*, 424. [CrossRef]

5. Uebe, J.; Paulauskiene, T.; Boikovych, K. Cost-effective and recyclable aerogels from cellulose acetate for oil spills clean-up. *Environ. Sci. Pollut. Res.* **2021**, *28*, 36551–36558. [[CrossRef](#)]
6. Paulauskiene, T.; Uebe, J.; Ziogas, M. Cellulose aerogel composites as oil sorbents and their regeneration. *PeerJ* **2021**, *9*, e11795. [[CrossRef](#)]
7. Paulauskiene, T.; Teresiute, A.; Uebe, J.; Tadzijevas, A. Sustainable Cross-Linkers for the Synthesis of Cellulose-Based Aerogels: Research and Application. *J. Mar. Sci. Eng.* **2022**, *10*, 491. [[CrossRef](#)]
8. Hüsing, N.; Schubert, U. Aerogels—Airy materials: Chemistry, structure and properties. *Angew. Chem. Int. Ed.* **1998**, *37*, 22–45. [[CrossRef](#)]
9. Li, Y.; Liu, X.; Cai, W.; Cao, Y.; Sun, Y.; Tan, F. Preparation of corn straw based spongy aerogel for spillage oil capture. *Korean J. Chem. Eng.* **2018**, *35*, 1119–1127. [[CrossRef](#)]
10. Virtanen, T.; Svedström, K.; Andersson, S.; Tervalu, L.; Torkkeli, M.; Knaapila, M.; Kotelnikova, N.; Maunu, S.L.; Serimaa, R. A physico-chemical characterisation of new raw materials for microcrystalline cellulose manufacturing. *Cellulose* **2012**, *19*, 219–235. [[CrossRef](#)]
11. Rajanna, S.K.; Kumar, D.; Vinjamur, M.; Mukhopadhyay, M. Silica Aerogel Microparticles from Rice Husk Ash for Drug Delivery. *Ind. Eng. Chem. Res.* **2015**, *54*, 949–956. [[CrossRef](#)]
12. Palma Rodríguez, H.M.; Delgado, R.S.; Olarte Paredes, A.; Salgado Delgado, A.M.; Hernández Uribe, J.P.; Pineda, E. Partial Characterization of Aerogels Made from Chayotextle and Potato Starch. *Emir. J. Food Agric.* **2022**, *34*, 634–641. [[CrossRef](#)]
13. Feng, J.; Nguyen, S.T.; Fan, Z.; Duong, H.M. Advanced fabrication and oil absorption properties of super-hydrophobic recycled cellulose aerogels. *Chem. Eng. J.* **2015**, *270*, 168–175. [[CrossRef](#)]
14. Liebner, F.; Potthast, A.; Rosenau, T.; Haimer, E.; Wendland, M. Cellulose aerogels: Highly porous, ultra-lightweight materials. *Holzforschung* **2008**, *62*, 129–135. [[CrossRef](#)]
15. Jiang, F.; Hsieh, Y.L. Amphiphilic superabsorbent cellulose nanofibril aerogels. *J. Mater. Chem. A* **2014**, *2*, 6337–6342. [[CrossRef](#)]
16. Bhandari, J.; Mishra, H.; Mishra, P.K.; Wimmer, R.; Ahmad, F.J.; Talegaonkar, S. Cellulose nanofiber aerogel as a promising biomaterial for customized oral drug delivery. *Int. J. Nanomed.* **2017**, *12*, 2021. [[CrossRef](#)]
17. Zaman, A.; Huang, F.; Jiang, M.; Wei, W.; Zhou, Z. Preparation, Properties, and Applications of Natural Cellulosic Aerogels: A Review. *Energy Built Environ.* **2020**, *1*, 60–76. [[CrossRef](#)]
18. Ferronato, N.; Torreta, V. Waste mismanagement in developing countries: A review of global issues. *Int. J. Environ. Res. Public Health* **2019**, *16*, 1060. [[CrossRef](#)]
19. Meng-Chuen Chen, D.; Bodirsky, B.L.; Krueger, T.; Mishra, A.; Popp, A. The world's growing municipal solid waste: Trends and impacts. *Environ. Res. Lett.* **2020**, *15*, 074021. [[CrossRef](#)]
20. Demirel Bayik, G.; Altin, A. Production of sorbent from paper industry solid waste for oil spill cleanup. *Mar. Pollut. Bull.* **2017**, *125*, 341–349. [[CrossRef](#)]
21. Korhonen, O.; Budtova, T. All-cellulose composite aerogels and cryogels. *Compos. Part A Appl. Sci. Manufact.* **2020**, *137*, 106027. [[CrossRef](#)]
22. Beluns, S.; Gaidukovs, S.; Platnieks, O.; Gaidukova, G.; Mierina, I.; Grase, L.; Starkova, O.; Brazdausks, P.; Kumar Thakur, V. From Wood and Hemp Biomass Wastes to Sustainable Nanocellulose Foams. *Ind. Crops Prod.* **2021**, *170*, 113780. [[CrossRef](#)]
23. Zhu, J.; Zhu, Y.; Ye, Y.; Qiu, Z.; Zhang, Y.; Yu, Z.; Sun, X.; Bressler, D.C.; Jiang, F. Superelastic and Ultralight Aerogel Assembled from Hemp Microfibers. *Adv. Funct. Mater.* **2023**, *33*, 2300893. [[CrossRef](#)]
24. Kaur, J.; Sharma, K.; Kaushik, A. Waste hemp-stalk derived nutrient encapsulated aerogels for slow release of fertilizers: A step towards sustainable agriculture. *J. Environ. Chem. Eng.* **2023**, *11*, 109582. [[CrossRef](#)]
25. Semlali Aouragh Hassani, F.-Z.; Hamid Salim, M.; Kassab, Z.; Sehaqui, H.; Ablouh, E.H.; Bouhfid, R.; El Kacem Quiss, A.; El Achaby, M. Crosslinked starch-coated cellulosic papers as alternative food-packaging materials. *RSC Adv.* **2022**, *12*, 8536–8546. [[CrossRef](#)] [[PubMed](#)]
26. Long, L.; Weng, Y.; Wang, Y. Cellulose Aerogels: Synthesis, Applications, and Prospects. *Polymers* **2018**, *10*, 623. [[CrossRef](#)] [[PubMed](#)]
27. Yan, C.; Wang, R.; Wan, J.; Zhang, Q.; Xue, S.; Wu, X.; Zhang, J.; Zhang, J.; Lu, Y.; Cong, W. Cellulose/microalgae composite films prepared in ionic liquids. *Algal Res.* **2016**, *20*, 135–141. [[CrossRef](#)]
28. Pereira, L.S.; Feitosa, J.P.; Morais, J.P.S.; de Freitas Rosa, M. Bacterial cellulose aerogels: Influence of oxidation and silanization on mechanical and absorption properties. *Carbohydr. Polym.* **2020**, *250*, 116927. [[CrossRef](#)]
29. Ahmed, Y.M.Z.; Ewais, E.M.M.; El-Sheikh, S.M. Potato starch consolidation of aqueous HA suspension. *J. Asian Ceram. Soc.* **2015**, *3*, 108–115. [[CrossRef](#)]
30. Aerogel.org. 2023. Available online: <https://www.aerogel.org/> (accessed on 5 June 2023).
31. Illera, D.; Mesa, J.; Gomez, H.; Maury, H. Cellulose Aerogels for Thermal Insulation in Buildings: Trends and Challenges. *Coatings* **2018**, *13*, 345. [[CrossRef](#)]
32. Chung, V.N.; Nguyen, T.S.; Huynh, K.P.H.; Chau, N.D.Q. Fabrication of Cellulose Aerogel from Waste Paper and Banana Peel for Water Treatment. *Chem. Eng. Trans.* **2022**, *97*, 337–342.

33. Nguyen, H.S.H.; Phan, H.H.; Huynh, H.K.P.; Nguyen, S.T.; Nguyen, V.T.T.; Phan, A.N. Understanding the effects of cellulose fibers from various pre-treated barley straw on properties of aerogels. *Fuel Process. Technol.* **2022**, *236*, 107425. [[CrossRef](#)]
34. Bigdoli, H.; Mortazavi, Y.; Khodadadi, A.A. A functionalized nano-structured cellulosic sorbent aerogel for oil spill cleanup: Synthesis and characterization. *J. Hazard. Mater.* **2019**, *366*, 229–239. [[CrossRef](#)]

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