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Sustainable Cross-Linkers for the Synthesis of Cellulose-Based Aerogels: Research and Application

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Abstract: Cellulose aerogels with polyester resin as cross-linkers have attracted much attention. This study describes the route to produce a fully bio-based aerogel with high added value from waste paper and starch, cellulose acetate and starch–cellulose acetate mixture as cross-linkers for oil adsorption, instead of the environmentally harmful polyester resin. The manufacturing process is simple, sustainable and cost-efficient, without releasing harmful by-products into the environment. The effects of different cross-linkers on the oil adsorption, dynamic oil retention, reusability and morphology of the aerogels were studied in detail. Experimental results show that these environmentally friendly recycled aerogels have a very low density, i.e., —0.0110–0.0209 g cm^{-3}, and highly porous structures, with a porosity of 96.74–99.18%. The synthesized hydrophobic aerogels showed contact angles of ∼124–129°. The compression moduli are lower than that of an aerogel with polyester as a cross-linker, but the compression modulus of the mixture of starch and cellulose acetate especially shows a higher value than expected. The sorption capacity of the aerogels with bio-based cross-linkers was significantly increased compared to the aerogels with polyester; it is now up to 56 times their own weight. The aerogels also have good oil-retention properties.

Keywords: cellulose aerogel; sorbent; paper waste; cross-linker; oil-spill cleanup; sorption capacity; oil/water separation; oil-retention ability; reusability

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1. Introduction

Even though we are moving toward a fossil-free economy, the world is still very dependent on oil, and there are numerous oil spills every year [1,2].

An oil spill is the release of liquid petroleum hydrocarbons into the environment, often to the marine ecosystem, due to incidents during oil exploration and production, oil loading operations or oil transportation. Oil spills not only change the color, acidity, taste and smell of water, but, most important, they have a toxic effect on organisms living in water. Oil spills at sea are usually much more damaging than those on land, as they can spread quickly and easily, forming a thin oil slick [1–4].

The problem of oil spills has never been so prominent for humanity than with the arising awareness of climate change and health. Thus, there is a need for sustainable innovative solutions which will help to minimize risks for the local ecosystem and negative impacts on human well-being.

There are a large number of oil-spill-containment techniques and affected-area-recovery methods, as well as cleanup equipment, that have been developed and implemented to respond to oil spills. Oil-spill-treatment methods can be systemized as mechanical, chemical or biological [4–7].

Aerogel is a gel in which the continuous liquid phase has been replaced by air through a specific drying process, resulting in a solid porous material [8,9]. The typical characteristics of an aerogel are high porosity (~99%), high surface area (>100 m^2 g^{-1}), and low density (0.003–0.500 g cm^{-3}) [8–13]. The main benefit of aerogels for oil-spill response is the recovery of the oil and the recyclability of the sorbent material [14]. Several different kinds of recovery methods have been developed, such as extraction, distillation and mechanical squeezing [15–17]. All of these characteristics are ideal for oil-sorption applications, allowing for rapid sorption and flotation on water [9,12,18,19].

Due to their unique biodegradability, biocompatibility, sustainability and regeneration, polysaccharides are ideal for use in the production of aerogels [20].

In order to enhance adsorption selectivity for nonpolar liquids from highly polar water, these bio-polysaccharide aerogels could be hydrophobized with organosilicon compounds, such as methyltrimethoxysilane (MTMS), hexadecyltrimethoxysilane (HDTMS) or perfluorooctyltriethoxysilane (PFOTES) [6,9,21–27].

A cross-linking agent can be defined as a substance that can cross-link molecules and improve the properties of aerogels. The use of a cross-linker in aerogels to form a 3D structure increases molecular weight; provides better mechanical properties; improves stability; and affects physical properties, such as polymer elasticity and insolubility [28].

One of the complex polymeric composites that can participate as a cross-linking agent in the synthesis of aerogels is a polyester polyol otherwise known as a polyester resin [29]. It is a synthetic material made from unsaturated organic dicarboxylic acids and polyhydric alcohols. Petroleum-based unsaturated polyester resins, due to their good mechanical properties, low density, low cost and easy processing, are one of the most widely used thermosetting polymers in the world. The polyester resin is conventionally produced from a petroleum-based reactive diluent styrene, together with a solid prepolymer, which is an unsaturated polyester having fumarate groups in its chain, or it might be the product of the esterification of an epoxy monomer with methacrylic acid.

However, there is a problem with the use of styrene in polyester resin, which is highly volatile and has been identified by the Environmental Protection Agency as a hazardous air pollutant [4]. In addition, the US Department of Health and Human Services has substantiated that it is a carcinogenic compound under the National Toxicology Program [30]. It is therefore very important to look for environmentally friendly alternatives that could serve as a substitute for polyester resin.

Cellulose acetate is the main component of cigarette butts, which are discarded all over the world without being recycled is a valuable polymer for numerous applications [31,32]. Cellulose acetate is a widely recognized biodegradable biopolymer in the scientific community that is derived from cellulose [33]. Cigarette butts are the most known variety of anthropogenic litter on Earth, as roughly 5.6 trillion cigarettes were used up around the world in 2010, and nine trillion are expected by 2025 [34]. Cellulose acetate–based sorbents have a good oil sorption capacity [35].

Starch, as a low-cost natural polymer, is widely used in paper, food, adhesive and many other industries [36,37]. Starch is an inexpensive biodegradable nutrient that can form an integrated gel network structure in the absence of other cross-linkers [20].

This paper presents the production of cellulose aerogel with high adsorption capacity after hydrophobization with MTMS (because of its simple applicability) for the three oil types (crude oil, marine diesel oil and lubricating oil) from paper waste. The problematic unsaturated polyester resin was replaced with biodegradable, sustainable cross-linkers, such as starch, cellulose acetate and a mixture thereof, and the application as sorbents for the three oils mentioned was tested.

2. Materials and Methods

2.1. Chemicals and 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”), MoTip polyester resin (MOTIP DUPLI Group, Wolvega, The Netherlands), cellulose acetate from cigarette-filters production waste (JSC Phillips Morris Baltic, Klaipeda, Lithuania) and potato starch (Aloja Starkelsen Ltd., Alojas lauku teritorija, Latvia) were used as raw materials. Methyltrimethoxysilan (MTMS of 98%; Sigma-Aldrich Chemie GmbH, Taufkirchen, Germany) was used for hydrophobization of aerogels. Acetone was purchased from JSC Labochema (Klaipeda, Lithuania).

For the investigation of 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 (Mazeikiai, 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 (Klaipeda, Lithuania), and the lubricating oil PEMCO iDrive 105 SAE 15w-40 with a density of 877 kg m^{-3} and a dynamic viscosity of 0.0038 Pa s from JSC SCT Lubricants (Klaipeda, Lithuania) were used.

2.2. Aerogels Synthesis Process

The main steps of aerogels synthesis are shown in Figure 1.

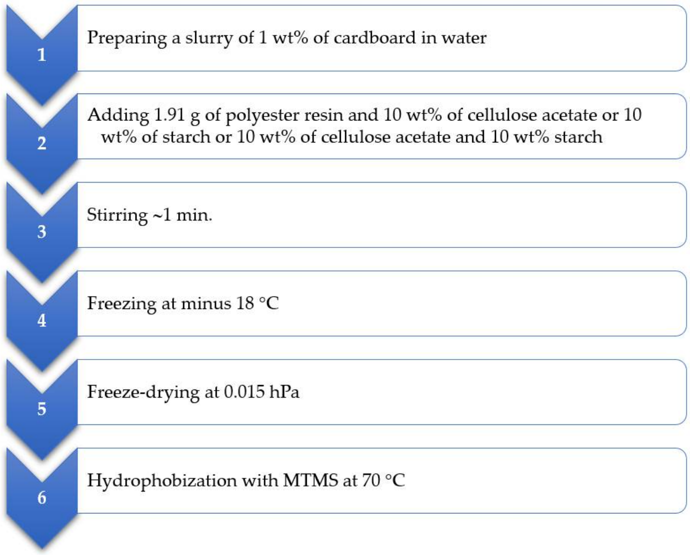

Figure 1. Aerogels synthesis scheme.

Cardboard was shredded up to 0.5 cm^2 in size, and 2 g of it in 200 mL distilled water (1 wt%) was homogenized 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). For the control samples, 1.91 g (1.11 wt%, ρ = 1.145 g cm^{-3}) of polyester resin was added to the mixture and homogenized for about one minute. Then the mixture was poured into 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, Lillerod, Denmark).

After freeze-drying, the samples were put in a 3 L beaker. A total of 1 g MTMS per 1 g aerogel was added to the bottom of the beaker in a 50 mL crystallizing dish and separated from the samples above with a plastic mesh. The top of the beaker was covered with aluminum foil and placed in the oven at 70 °C for 12 h.

The samples with cellulose acetate as a cross-linking agent were prepared analogously to control samples. Instead of a polyester resin, 0.2 g cellulose acetate dissolved in 10 mL acetone was used so that the ration of cellulose acetate to paper waste was 1 to 10. The cigarette filters were crushed, weighed and dissolved in acetone, with swirling.

In the case with starch as a cross-linking agent, 0.2 g starch was dissolved in 20 mL hot water.

Finally, in the case of the mixture of starch and cellulose acetate as a cross-linking agent, the cellulose mixture was mixed with a starch solution of 0.1 g starch in 10 mL hot water, as before described, and homogenized for about one minute. Then a cellulose acetate of 0.1 g cellulose acetate in 5 mL acetone, as formerly described, was added, and the mixture was mixed for another minute.

The photographs of the synthesized aerogels with different cross-linkers are shown in Figure 2.

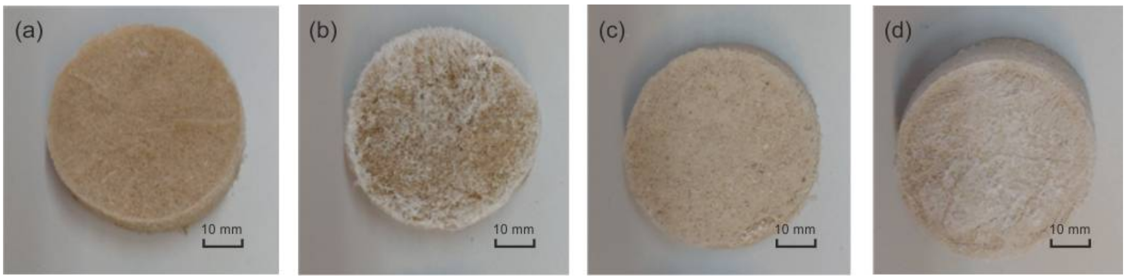

Figure 2. Photographs of aerogels with different cross-linking agents: (**a**) control samples (polyester resin), (**b**) cellulose acetate, (**c**) starch and (**d**) starch and cellulose acetate.

To hydrophobize the samples [29,38], MTMS (1 g per 1 g aerogel) was added to the bottom of a sufficiently large glass container in a small crystallizing dish, and the samples were arranged vertically above it, separated with a plastic net. The glass container was covered on top with aluminum foil and placed in the oven at 70 °C for 12 h.

2.3. Investigation of Aerogel Properties

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 was calculated as given in Reference [38]:

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

where ρ_s is the bulk density of the aerogel samples, and $\rho_{t.s.}$ is the theoretically density of the mixtures of the different cross-linkers and the cellulose of the paper waste and can be calculated according to the following equation [39]:

$$\rho_{t.s.} = \frac{1}{\frac{w_{cellulose}}{\rho_{cellulose}} + \frac{w_{cross\text{-}linker}}{\rho_{cross\text{-}linker}}} \tag{2}$$

where $w_{cellulose}$ and $w_{cross\text{-}linker}$ are the parts of cellulose and cross-linker in the sample; $\rho_{cellulose}$ and $\rho_{cross\text{-}linker}$ are the densities of the cellulose and cross-linker. According to the literature, the density of cellulose is 1.5 g cm^{-3} [40], cellulose acetate is 1.3 g cm^{-3} [41], starch is 1.45 g cm^{-3} [42] and the density of the polymer resin is given in Section 2.2.

The wettability of the aerogel surface was investigated by measuring the water contact angle by using the sessile drop method. A small drop of water is placed on the surface of aerogel, and a high-resolution camera from a smartphone is used for a close-up photograph. ImageMeter tool measures the angle of an image and determines the degree of hydrophobicity of aerogels.

The morphology and structure of the aerogels were characterized by using scanning electron microscopy (SEM) S-3400N, Hitachi (Hitachi, Japan). The elemental composition of the aerogels was estimated by energy-dispersive X-ray spectroscopy (EDS), using a Bruker Quad 5040 spectrometer, Hitachi (Hitachi, Japan). The samples were placed on an aluminum holder and not sputtered.

The compressive strength of the aerogels was determined by applying the Zwick/Roell Z020 with testXpert II testing software (ZwickRoell GmbH & Co, Northeim, Germany) with the strain rate of 1 mm min^{-1}. The modulus of elasticity is defined as the value of the slope of the linear elastic region that occurs at low compression. Perfect plane parallelism of the aerogel specimens cannot be realized for practical reasons, and this is why the compressive stress curves do not start out linear, but are mostly parabolic, and only show linearity from a compression of >1%. For this reason, the slope is determined at higher compression, in the range of 30% (first point) and 70% (second point).

The maximum sorption capacity of aerogels (Q) was determined by using Reference [38]. For this, the aerogel samples were weighed before measurement. Then the aerogel samples were immersed in oil for 5 min. The samples were then removed from the oil, allowed to drain for 1 min and weighed again:

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

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

Due to the intended use as sorption material for oil pollution, it is necessary to increase its hydrophobicity with MTMS. The contact angles of water droplets were measured at several locations, and the results were averaged.

Hydrophobicity test results of aerogels with different cross-linking agents are presented in Figure 3 and Table 1.

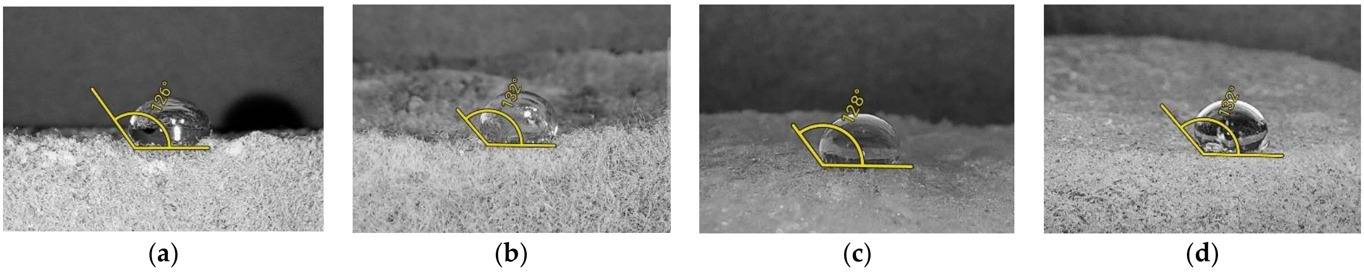

Figure 3. Photographs of an aerogels with different cross-linking agents and the biggest contact angles of water droplets: (**a**) control samples (polyester resin), (**b**) cellulose acetate, (**c**) starch and (**d**) starch and cellulose acetate.

Table 1. Hydrophobicity test results of aerogels.

	Cross-Linking Agent			
	Polyester Resin (Control Samples)	**Cellulose Acetate**	**Starch**	**Starch–Cellulose Acetate**
Contact angles of water droplets, °	124 ± 2	128 ± 4	127 ± 2	129 ± 2

Compared with the control samples, the contact angles were at least 124, so that the same effect on hydrophobicity by the modification technique could be observed on the aerogel samples. Furthermore, it can be concluded that all manufactured aerogels are hydrophobic because their contact angles of water droplets are greater than 90° [43,44].

Since starch has a slightly higher density than cellulose acetate, the porosity of the corresponding aerogel samples calculated from the densities is also somewhat lower than that of the aerogel samples with cellulose acetate as the cross-linker (Table 2).

Table 2. Densities and calculated porosities of the aerogel samples.

Aerogel Sample	**Density in g cm^{-3}**	**Porosity in %**
Control	0.021 ± 0.001	96.74
Starch	0.018 ± 0.001	98.68
Starch–cellulose acetate	0.014 ± 0.001	98.84
Cellulose acetate	0.011 ± 0.001	99.18

3.2. Morphology

Changes in the morphological structure of the aerogels were observed visually and by scanning electron microscopy before and after MTMS coating. The porous surfaces were observed before and after modification to detect changes in the structure of the aerogels by silanization process and using different cross-linking agents. Successful silanization of the cellulose aerogel was confirmed by energy-dispersive spectrometry spectra same, as was performed by Reference [45].

When studying the silicon content in at% of the different samples of Table 3, that the increase in silicon content is very small in the case of the cellulose aerogel sample with cellulose acetate as a cross-linker. It is discussed with the content in at%, as this corresponds to the atomic percentage as the number of atoms of the respective element calculated from the weight fraction in wt% of the EDS spectra. In comparison, the Si content of the sample with starch as a cross-linker increases by a factor of almost three. Since both starch and cellulose consist of glucose with up to three free silanizable OH groups per glucose unit, the comparatively high increase in silicon in Table 3 can be explained. In the case of cellulose acetate as a cross-linker, only about one free OH group is present on average. Since the underlying cellulose fiber itself still has up to three free OH groups per glucose unit, the small increase in the Si content could be explained by partial coverage of the cellulose fiber by cellulose acetate. This could also be assumed in the case of the aerogel with both types of cross-linkers. Since the cellulose suspension is first mixed with starch and then with cellulose acetate, the cellulose acetate could cause partial coverage of cellulose and starch and thus conceal part of the free OH groups.

Table 3. Extracted elemental composition from the energy-dispersive X-ray spectra of the aerogels.

Cross-Linking Agent	Modification with MTMS	Dimension	Chemical Element			
			C	O	Si	Other
Control samples with polyester resin	No	wt%	42.24	54.78	0.45	2.53
		at%	50.07	48.75	0.23	0.95
	Yes	wt%	38.28	56.52	0.82	4.38
		at%	46.38	51.41	0.42	1.79
Cellulose acetate	No	wt%	33.53	57.38	0.91	8.18
		at%	41.98	53.93	0.48	3.61
	Yes	wt%	33.86	58.82	0.95	6.37
		at%	42.05	54.84	0.51	2.60
Starch	No	wt%	33.82	60.92	0.25	5.01
		at%	41.61	56.26	0.13	2.00
	Yes	wt%	33.11	61.33	0.67	4.89
		at%	40.86	56.82	0.35	1.97
Starch–cellulose acetate	No	wt%	35.19	58.77	0.62	5.42
		at%	43.20	54.16	0.33	2.31
	Yes	wt%	34.42	58.09	1.13	6.36
		at%	42.57	53.93	0.60	2.90

In the SEM images shown in Figure 4, we can see the diameter of the filaments, which varies from 15.5 to 41.2 µm, as well as their arrangement, which determines the pore size distribution. As in the case of Reference [46], the granular appearance on the surface could indicate the MTMS coating of the individual filaments. The edges of the filaments between the comparatively large cellulose fibers could be due to the respective cross-linkers, indicating the formation of cross-links. Long filaments are parts of shredded cardboard, while chips and fibers are cross-linking agents, and granules are MTMS. Overall, the photos of the aerogel samples show a high degree of porosity in the structure, which consists of an interconnected network and a distribution of cellulose and cross-linking fibers.

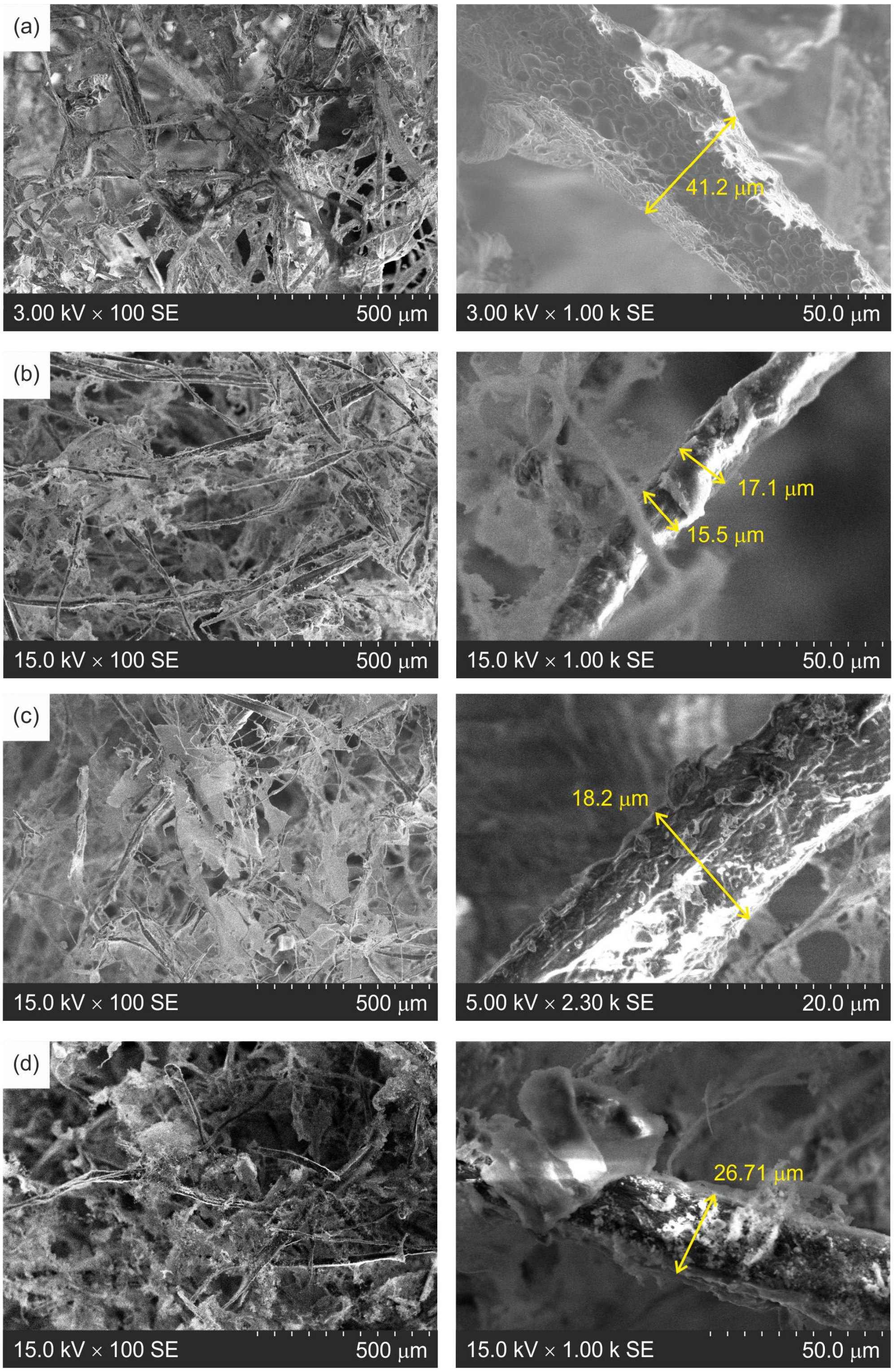

Figure 4. Microscopic structure of the aerogels with a different cross-linkers (left photo shows the overview before modification with MTMS and the right photo show a detail in 10-fold-higher magnification than the left photo after modification with MTMS): (**a**) polyester resin (control), (**b**) cellulose acetate, (**c**) starch and (**d**) starch–cellulose acetate.

3.3. Results of the Compressive Mechanical Properties Test

The average compression moduli of the four types of aerogel samples are 34,166 Pa for the control sample, 20,881 Pa for the aerogel samples with starch, 15,427 Pa for the aerogel samples with the mixture of starch and cellulose acetate and 2168 Pa for the aerogel samples with cellulose acetate as a cross-linker (Figure 5). As in the literature [47], the compression moduli can be correlated linearly—proportional to the density and inversely proportional to the porosity calculated from it.

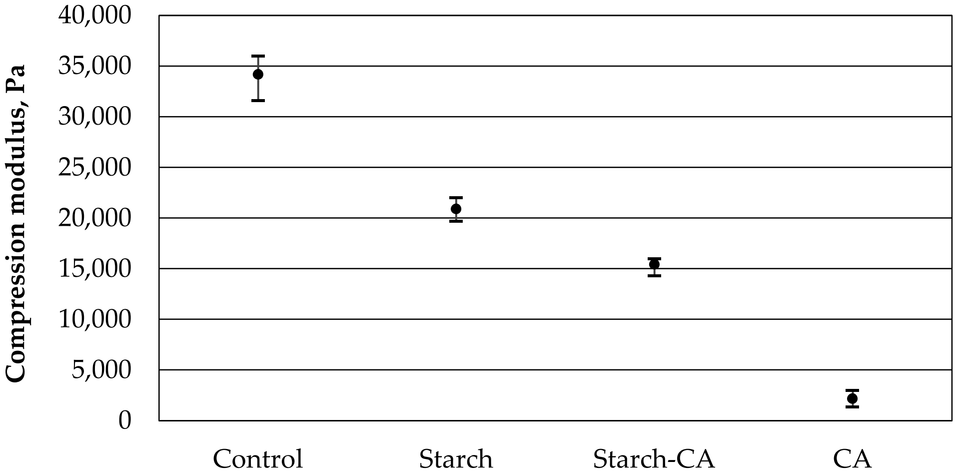

Figure 5. Compression modulus of the different aerogel samples.

To compare the stiffnesses of the different specimens, the quotient of the compression modulus and the respective specimen density is used. A value of 1.6 MPa cm^3 g^{-1} is obtained for the control specimens, and 1.2, 1.1 and 0.2 MPa cm^3 g^{-1} are obtained for the specimens with starch, with the mixture of starch and cellulose acetate and with cellulose acetate alone, respectively. It can be seen from these values that the aerogel specimens with starch and with the starch–cellulose acetate blend have very similar stiffnesses, while the stiffness of cellulose acetate cross-linked specimens is comparatively low. Starch in the blend appears to offset the detrimental effect of cellulose acetate on the stiffness of the aerogel sample. Furthermore, it can be seen that the loss in stiffness when using only 10 wt% starch as a cross-linker is rather small compared to the control sample. The control sample has the same amount of cross-linker as cellulose content in the sample.

The values of the stiffness itself are rather low compared to aerogel samples from chemical gelation, as in Reference [47], and the cross-linkers from starch, and they might be a result of the non-chemical formation process [48].

3.4. Maximum Sorption Capacity

The sorption capacity is an important parameter that determines the maximum amount of oil or its products that can be sorbed per gram of aerogel.

From Figure 6, it can be seen that the lowest sorption capacity for the types of oil studied is for the control samples with polyester resin as the cross-linking agent, while the highest sorption capacity is for starch–cellulose acetate aerogels. For this sample in particular, it can be assumed that either the actual porosity is probably higher than the calculated one in Section 3.1 or the cellulose acetate acts as an independent sorbent analogous to [35]. This suggests itself, since cellulose acetate is inherently hydrophobic [35,49].

3.5. The Regeneration of the Aerogel Samples by Squeezing

The data on the reuse of aerogels after regeneration are very important to determine whether the product is effective in further sorption after squeezing.

The sorption-regeneration process was performed four times because it was observed that the volume of the aerogels changes after the first cycle; they begin to rupture after the fourth cycle; and the sorption capacity remains constant during the second, third and fourth cycles.

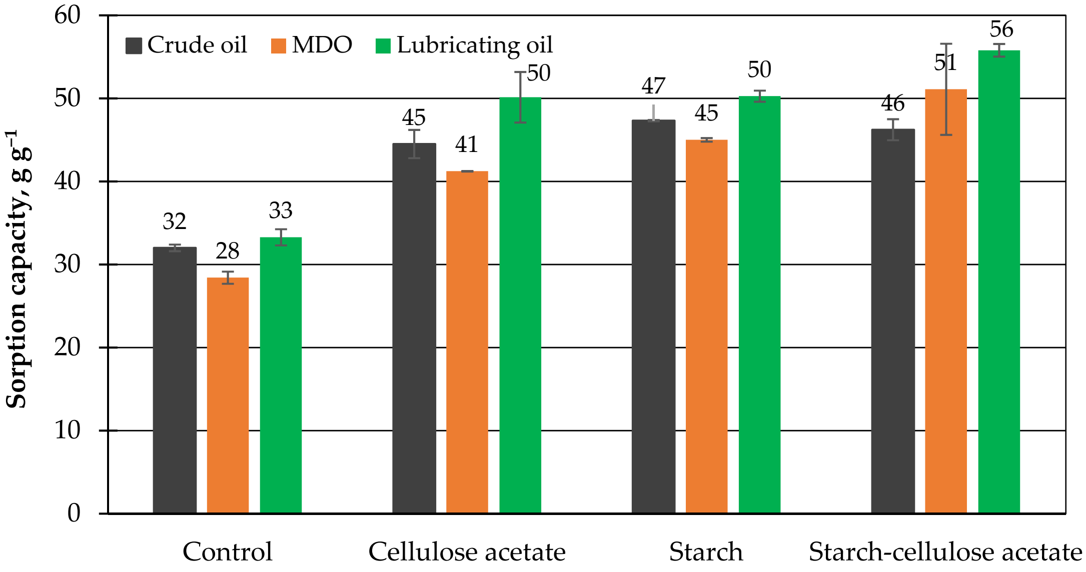

Figure 6. Results of aerogels sorption capacity to crude oil, MDO and lubricating oil.

When the aerogel samples were first used as sorbents, the sample with starch as a cross-linker had the highest sorption capacity of 47 ± 0.08 g g^{-1} (Figure 7). The sorption capacities after the second, third and fourth sorption-regeneration cycles were significantly lower at about 6 g g^{-1}. The efficiency of these aerogels (the first point is taken as a reference point and set on 100%) was reduced by 87% after the first sorption-regeneration cycle.

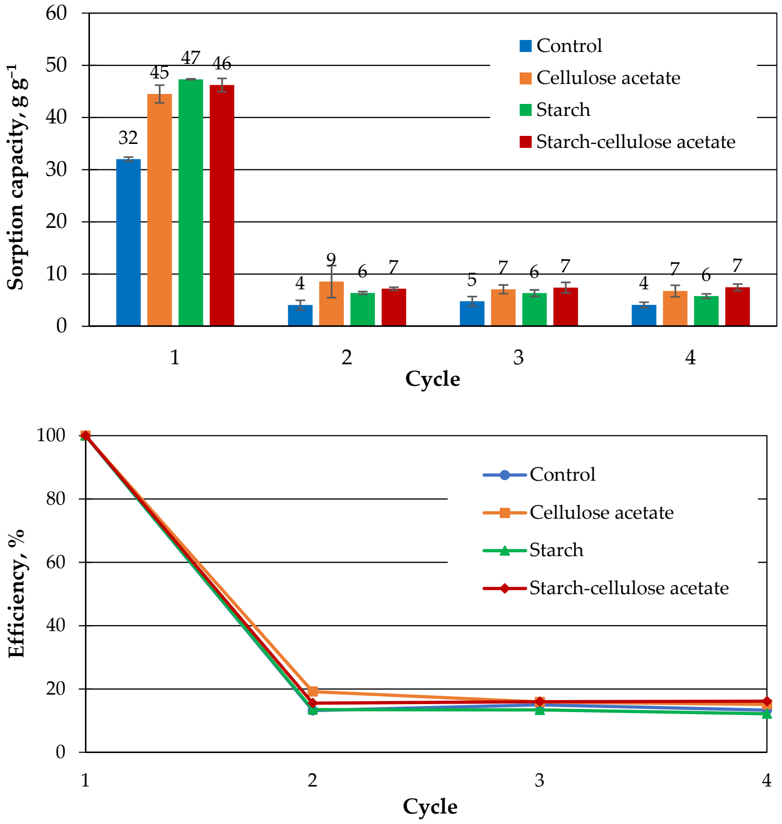

Figure 7. Results of the aerogels with different cross-linkers maximum crude oil-sorption capacity (**upper figure**) and reuse efficiency of the samples on crude oil by each cycle (**lower figure**).

The aerogels with cellulose acetate as a cross-linking agent had a slightly lower sorption capacity of 45 ± 1.70 g g^{-1} than the former starch-linked aerogels. However, their sorption capacity was higher in the second sorption-regeneration cycle. The efficiency was reduced by 81%. In the third and fourth cycles, the sorption capacities of the aerogels were about 7 g g^{-1} regardless of the cross-linking agent.

When comparing these results of the sorption capacity of crude oil of the aerogels with starch, cellulose acetate and starch–cellulose acetate as cross-linking agents with those of the control samples, an increase of 15 ± 1 g g^{-1} can be seen. However, the loss of sorption efficiency for all of these aerogels was about 87%. This could be explained similarly to References [29,49], according to which the destruction of the pores and the inability of the samples to sorb more oil product would be responsible.

The aerogel samples with starch cellulose acetate as a cross-linker show the highest sorption capacity of 51 ± 5.49 g g^{-1} for marine diesel oil (Figure 8). However, it shows a ranking with the highest sorption capacity of the cross-linker mixture of starch cellulose acetate, followed by starch, cellulose acetate and lastly the control sample. The same ranking can be seen in the first sorption cycle. Interestingly, in this case, the cross-linker mixture does not seem to be simply a mixture of the individual components, but there is probably a mutually reinforcing effect between the two substances. However, this effect is no longer visible in the third and fourth sorption-regeneration cycles.

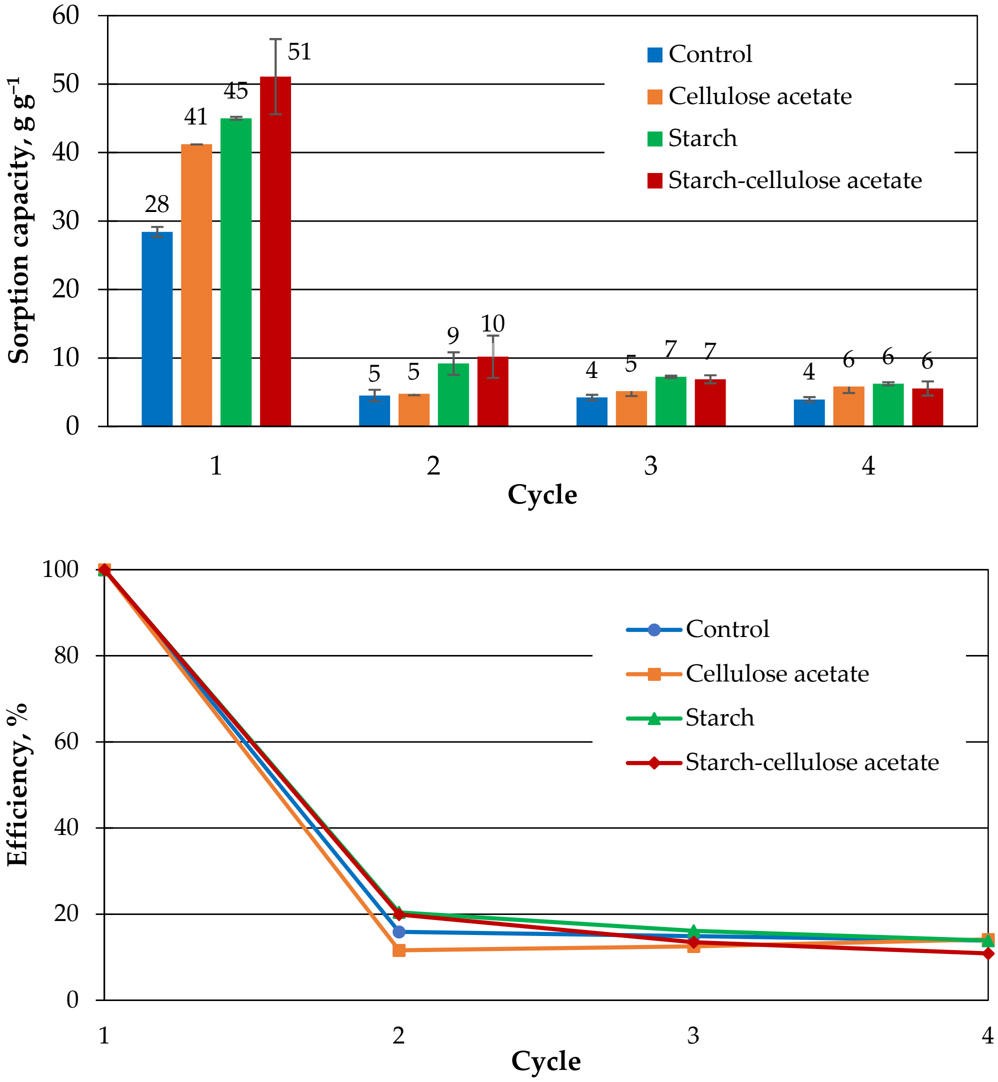

Figure 8. Results of the aerogels with different cross-linkers maximum MDO sorption capacity (**upper figure**) and reuse efficiency of the samples on MDO by each cycle (**lower figure**).

The sorption capacities of aerogels with starch, cellulose acetate and starch–cellulose acetate as cross-linking agents show higher capacity in marine diesel oil than in crude oil, but the efficiency loss is also about 88%. Similarly, the values of sorption capacities for the control sample are the lowest in the first and second sorption cycles.

As in the previous case, when a starch–cellulose acetate mixture was used as the cross-linking agent for the aerogels, the highest sorption capacity was found to be 56 ± 0.76 g g^{-1} (Figure 9). However, compared to the previous two oils, the aerogel samples with starch, cellulose acetate and their mixture also show significantly higher sorption capacity than the control sample in the second, third and fourth sorption-regeneration cycles. The efficiency loss of the aerogel samples with these cross-linkers was less than 80%, namely about 75%, for the sorption of lubricating oil.

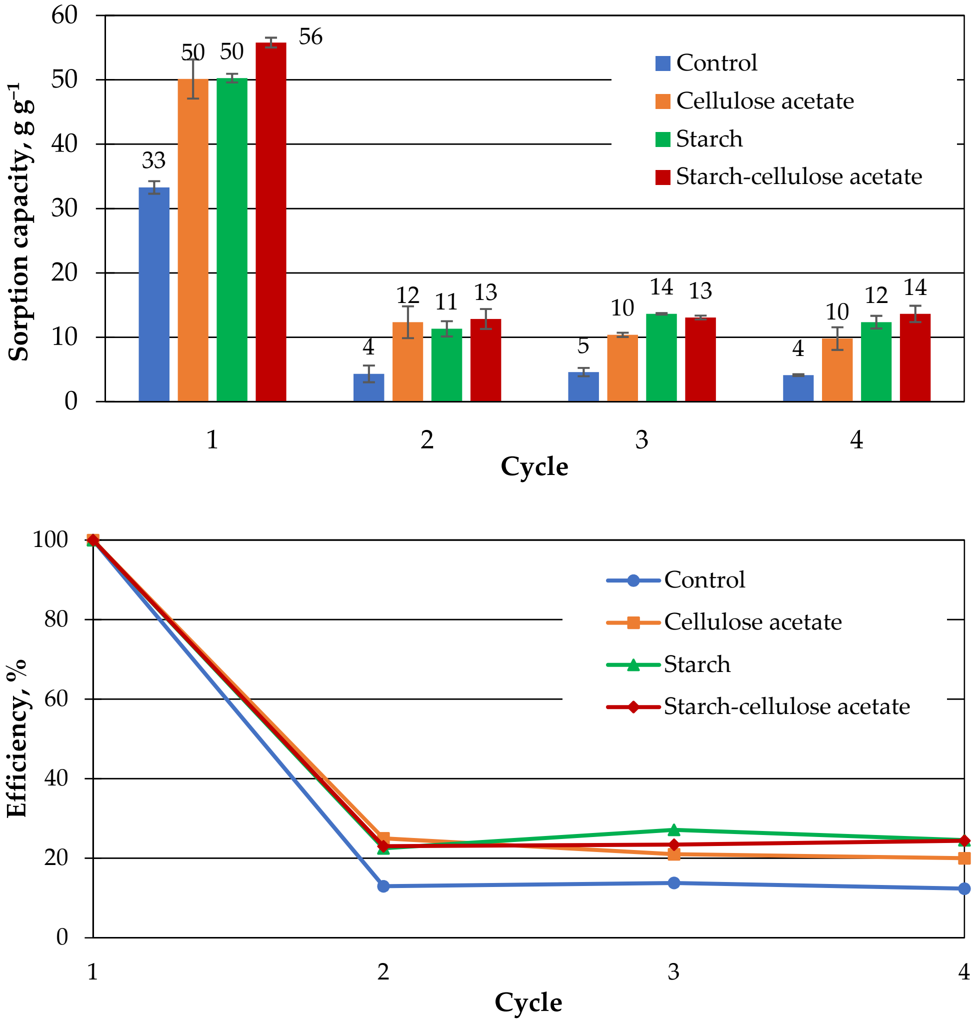

Figure 9. Results of the aerogels with different cross-linkers maximum lubricating oil sorption capacity (**upper figure**) and reuse efficiency of the samples on lubricating oil by each cycle (**lower figure**).

4. Conclusions

The results of the elemental composition measurement study showed an increase in the silicon content. The silicon content increase depends on the OH groups available. Cellulose cross-linked and presumably covered with cellulose acetate exhibits the lowest Si content increase, but it is still hydrophobic, as shown by the contact angle measurement and ability to sorption of oils. This analysis also showed that aerogels have an uneven, porous structure that is composed of filaments of different diameters.

The results of the mechanical properties test showed that the new aerogels have lower elasticity and stability than the control sample; thus, they have to be improved in the future for the possibility to reuse samples. The mixture of equal parts of starch and cellulose acetate as cross-linkers has a higher compression modulus than would be expected from a linear combination. This is clearly shown by the very similar specific compression moduli of the cross-linker mixture with that of the singular cross-linker of starch.

It was found that aerogels with a starch as a cross-linking agent had the highest sorption capacity of crude oil, i.e., 47 g g^{-1}, better performance of marine diesel oil, at 51 g g^{-1}; and lubricating oil at 56 g g^{-1} sorption capacity had aerogels with a mixture of starch and cellulose acetate as a cross-linking agent. Meanwhile, control samples with polyester resin as a cross-linking agent showed the lowest sorption capacity—up to 33 g g^{-1}.

It can be noted, that the aerogels reached a high sorption capacity at the beginning of the sorption-regeneration process, but decreased significantly during the subsequent sorption-regeneration cycles. During these sorption-regeneration cycles, an increase in the effect of the mixture of starch and cellulose acetate as the cross-linking agent was partially observed.

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