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[Tatiana B. Medvedeva](#)<sup>\*</sup>, Xeniiia S. Cherepanova, Valentin N. Parmon, [Nikolay V. Gromov](#)<sup>\*</sup>

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Review

# Production of Cellulose Materials from Oat Agricultural Wastes: A Review of Recent Approaches

Tatiana B. Medvedeva \*, Xeniiia S. Cherepanova, Valentin N. Parmon and Nikolay V. Gromov \*

Boreskov Institute of Catalysis; Lavrentiev av. 5, 630090 Novosibirsk, Russia

\* Correspondence: tanmedvedeva@catalysis.ru; gromov@catalysis.ru;

**Abstract:** The current trends in environment protection and sustainable development demand the investigations in the field of searching new perspective sources for any production. Oat husks seems to be a perspective resource. Cellulose isolated from oat wastes can be used for the production of different materials such as hydro and aerogels, bacterial, nanocrystal and nanofibrillated cellulose, cellulose based composites and ethers. All these materials can be spent in different industries: production of pharmaceuticals, medicine (including targeted delivery of drugs, sponges etc.), agriculture (controlled delivery of pesticides or fertilizers), electronics, production of films, membranes, plastics, coatings, paints, biodegradable packages. In this review, we will demonstrate prospects for using oat husks to obtain cellulose and for obtaining materials based on the main oat husk polysaccharide. This review will evaluate the main achievements in this area. The analysis will allow a reader to form an analytical opinion about the prospects for using oat husks as a source for obtaining valuable products.

**Keywords:** cellulose; nanocellulose; bacterial cellulose; nitrocellulose; oat husks; oat hulls; biomass valorization

## 1. Introduction

Nowadays humankind occurs a significant negative influence on the environment due to an extensive economic activity. However, it should be emphasized that an industrial production of any goods shall not be reduced in order of sustainable development of the society. Currently, there is an increasing pressure on the environment, and society's demands for materials are growing. Because of that, the technologies for processing plant renewable raw materials are of critical importance.

Cellulose is one of the most incredible natural polymers which can be assumed as most perspective substrate for the production of biobased materials. There are 240 billion tons of biomass is synthesized annually in the nature. Taking into account the average content of cellulose in plant raw materials equal to 20-45%, the annual growth of cellulose in the world reaches 100 billion tons. Given that cellulose is the main component of plant biomass, it makes cellulose the most abundant plant biopolymer on Earth. The widespread of cellulose and large volumes of its production open up prospects for its use as a raw material for both chemical synthesis and the production of new types of materials [1–3].

Cellulose can be derived from the numerous kinds of plant raw materials [4]. Perspective sources of cellulose can be divided into several groups.

1. Wastes of agriculture, most abundant ones are wheat straw, beet pulp, sugar cane pulp, corn cobs, grain husks.
2. Wastes of wood, paper industries.
3. Energy crops such as maize or sweet sorghum and herbaceous, miscanthus (*Miscanthus giganteus*), switchgrass (*Panicum virgatum*), kenaf (*Hibiscus cannabinus*), reed canary grass (*Phalaris arundinacea*) or giant reed (*Arundo donax*), and eucalyptus (*Eucalyptus globulus*) [5,6].

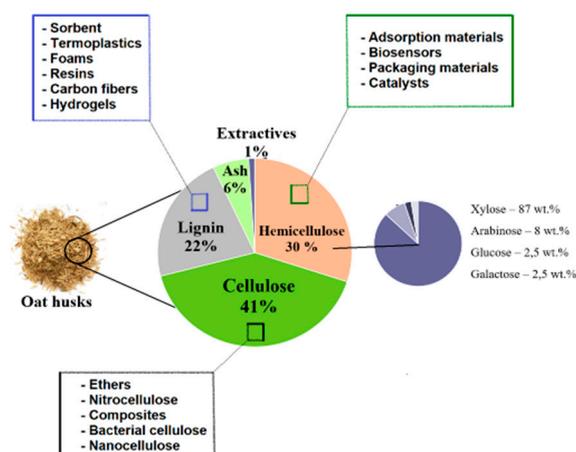
From these points of view, an attention can be paid to the prospects for processing oat husk (oat hull) agricultural wastes to produce cellulose and cellulose-based materials. A world production of

and processing oats (*Avena sativa*) was around 8 million, wherein the husks are removed, generating approximately 4 million tons of hulls, corresponding up to 50% of grain weight [7]. Nowadays, wastes of oat grain peeling are almost out of transformation and are stored in dumps, that take significant areas. Oat hulls contain approximately 90 g of fiber per 100 g of the feedstock [8]. It is higher than that of wheat (47 g/100 g) or corn bran (62 g/100 g) [8]. Oat hulls are mainly composed of lignocellulose and contains three polymer structures: cellulose, hemicelluloses and lignin which are presented as a complex net with plenty of interconnections [9]. Cellulose content in oat hulls could varies from 16-26% [9] to 30% [10] and even 35-45% [11]. Along with the glucose polysaccharide, the waste under consideration contains hemicelluloses (24–35%), and lignin (13–25%) [9,11].

Thus, due to features of production and storage as well as chemical composition, oat husk can be assumed as a valuable raw material for the production of biofuels [12,13] (for example, hydrochar), basic biopolymers (cellulose, hemicellulose and lignin) [14,15], and valuable chemicals such as monosaccharides [16], furan derivatives [17,18], organic acids [19], alcohols [20–22], phenols [23,24] and plenty of others (Figure 1).

Biopolymers (cellulose, hemicelluloses, lignin) derived from oat husks have wide application prospects. Based on lignin, sorbents, thermoplastics [13,15,25], polyurethane foam [26], phenol-formaldehyde resins, carbon fibers, hydrogels [15] can be produced. Hemicelluloses are also perspective for the production of biobased functional materials being suitable for adsorption [27], biosensing [28], packaging [29,30], catalysis and electrochemical applications [31] in food, medical, and energy storage industries [32]. And cellulose which is the main component of oat husks also has a lot of benefits as a substrate for the production of materials [8].

Thus, we can note: 1) high prospects for cellulose for obtaining materials based on it, 2) prospects for using oat husks to obtain this biopolymer. An analysis of the literature shows the existence of works in this area. However, to our knowledge, at present there are no review materials that bring together the prospects for using oat husks to obtain cellulose and materials based on it. This review will evaluate the main achievements in this area. The analysis will allow a reader to form an analytical opinion about the prospects for using oat husks as a source for obtaining valuable products.



**Figure 1.** Oat husk composition and perspective ways of its main component transformation to materials.

## 2. Composition of lignocellulose of oat hulls and the main challenges of its processing

Cellulose is well known to be a linear polymer which consists of D-glucose units (glucan units) connected by  $\beta$ -1,4-glycosite bonds [33]. The cellulose units are arranged in microfibrils which contains both amorphous and crystal domains [33]. Plenty of hydrogen bonds formed between glucan units of a chain and units of different chains makes cellulose quite robust for any treatment and provides high mechanical stability of lignocellulose structure [33]. As mentioned above, cellulose

content in oat hulls could reach 45% [11] and seems to depend on growing climate conditions [9]. High cellulose content makes oat hulls (oat husks) a very promising resource for the production of cellulose-based materials (Figure 1). Along with the glucose polysaccharide, the waste under consideration contains hemicelluloses (24-35%) and lignin (13-25%) [9,11,34,35]. Hemicelluloses, which are branched structures, are mainly presented by the saccharide polymers xylose, arabinose, and galactose. Xylan is the main hemicellulose polymer of oat hulls [9]. The overall strategy for the production of cellulose materials demands removal of hemicelluloses and lignin. From this point of view, the valorization of oat hulls meets several challenges due to the composite structure of biomass.

**Cellulose purity.** Cellulose, hemicelluloses and lignin in oat hull lignocellulose are interconnected via plenty of various linkages and bonds. This complex structure makes lignocellulose very resistant to any chemical and biological treatment [9,33,36]. In order to remove non-cellulosic components, a pretreatment (delignification) of oat husks has to be carried out [37,38]. Selecting the type of processing and its conditions is quite challenging. The cellulose structure and the amount of impurities affect the way of application of the resulting materials (Table 1).

**Table 1.** Perspective applications of celluloses derived from oat husk depending on the structural parameters of cellulose.

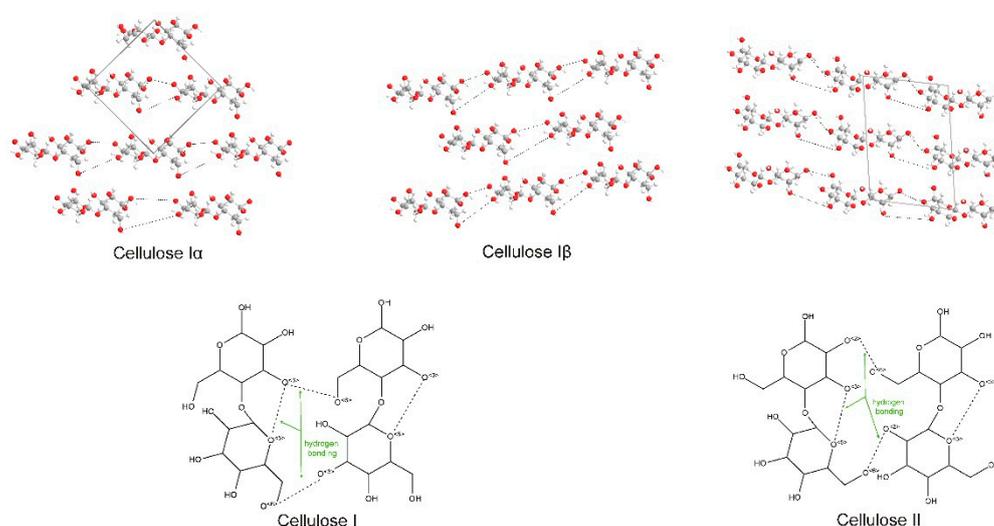
№	Method of pretreatment	Residual lignin, %	Residual hemicellulose, %	Nanocellulose			Composites	Nitrocellulose	Cellulose ethers	Hydrogels	Ref.
				Bacterial cellulose	Nanocrystal cellulose	Cellulose nanofibrils					
1	Acid only	7.2-12.1	6.9-7.4	+					+		[39]
		3.0-3.5	8			+			+		[40,41]
2	Acid+H <sub>2</sub> O <sub>2</sub>	3	7-8						+		[41,42]
3	Alkali only	3.3-4	8.1-9.4	+							[39]
4	Acid-alkali	0.32-0.8	0.7-3	+				+	+		[39,43-47]
5	Multi alkali-acid	1	1.4-26	+		+	+		+	+	[39,48,49]
6	Multi acid-alkali	< 1-2%	2-6.5	+							[39]
7	Alkali peroxide and physical methods	7.7-9.5	3-17				+				[50-52]
8	Alkali-acid combined with physical methods	7.2-12.6	3-26	+		+					[53]
9	Hydrotropic method	5-9	-						+		[54]

To improve the purity, cellulose undergoes bleaching. High purity of cellulose is of critical importance for some biomaterials. Thus, during the biomass processing, the delignification and bleaching is almost unattainable. On the other hand, degree of the delignification of the biomass causes a loss of cellulose product yield due to partial hydrolysis and solubilization of the polysaccharide during a treatment. An importance of purity of cellulose isolated from oat husks can be demonstrated via the following examples. Thus, 2.5% of lignin amount in cellulose decreases 5-hydroxymethylfurfural yield by the factor of 1.5 compared to one achieved from pure cellulose [55].

For nanocellulose to be produced, almost total purification of cellulose from lignin impurities is required [56,57]. During the synthesis of bacterial cellulose of a hydrolyzate obtained from the enzymatic hydrolysis of pretreated oat husks, lignin acts as an inhibitor of the biosynthesis and blocks the activity of enzymes or microorganisms [58]. On the other hand, upto 13% of lignin is appropriate for oat husk cellulose to be transformed to bioethanol [59].

**Wastewaters.** Another challenge of delignification of plant materials is the formation of wastewaters. However, a modern researches [60] show the reusability of a delignification agents as well as the use of environmentally friendly reagents [61].

**Structure.** Depending on the purpose of cellulose using, structural parameters are of critical importance. Besides purity (amount of residual lignin), crystallinity index (CI), crystallite size (determined by CSR) and degree of polymerization (DP) seems to be key parameters. These settings affect the order of cellulose structure, and hence the properties of the resulting material. It is well known that there are three main types of cellulose structure: triclinic I $\alpha$ , monoclinic I and cellulose II (Figure 2) [62].



**Figure 2.** Three main structures of cellulose.

For example, the structure of bacterial cellulose corresponds to the triclinic modification I $\alpha$  [63,64], and the structure of plant celluloses is usually cellulose I $\beta$  [64–67]. Plant cellulose has a lower crystallinity index compared to one of bacterial cellulose [66,68]. Nanocellulose (bacterial and nanocrystal) and bacterial celluloses have a high crystallinity index and degree of polymerization [69]. Some bacterial nanocellulose fibers have the values of Young's modulus and tensile strength of 10-65 GPa and 80-800 MPa, respectively [70]. These properties allow bacterial nanocellulose demonstrates advanced mechanical properties. On the other hand, for the production of bioethanol, organic acids, and furan derivatives from cellulose, the process is positively affected by decreasing the degree of polymerization and the crystallinity index. Such cellulose is better transformed by chemical catalysis or fermentation, since the lower density, better accessibility of cellulose chains for catalysts or enzymes and an existence of amorphous domains simplify the process.

Thus, it is necessary to pretreat the oat husk biomass in most cases to obtain cellulose based materials or valuable chemicals. Important characteristics that will determine the purpose of the isolated cellulose are impurities of noncellulose components, the degree of crystallinity and polymerization of cellulose.

### 3. Pretreatment and delignification of oat husks

As mentioned above, pretreatment and delignification are important stages of the production cellulose and cellulose based materials when transforming oat husks. Preliminary processing can be

carried out in the presence of different chemical agents: mineral and organic acid, alkalis as well as their combinations. Physical activation methods are also considered [33,71]. As the method of preprocessing raw materials affects the characteristics of the resulting biopolymers, a technique should be chosen carefully (Table 1). Selecting the pretreatment method is also important for deep transformation of cellulose. It is shown in [72] that the conditions of pretreatment of oat husks affect the productivity of ethylene. Thus, alkaline pretreatment of agricultural waste leads to a decrease in ethylene yield by 20% compared to a cellulose-containing sample obtained by the treatment with nitric acid.

Therefore, the pretreatment of oat husk biomass is the key to its subsequent transformation into the final product. The right approach will maximize the efficiency of product production and achieve the best cost and benefit ratio.

### 3.1. Acid, alkaly and physical treatment

#### 3.1.1. Acid treatment

Pretreatment by acids seems to be the most widely applied method for the transformation of lignocellulose biomass [73]. Pretreatment and delignification of oat husks by diluted solutions of sulfuric [73–77], nitric [11,39,78–82], hydrochloric [73], peracetic [40] acid is described.

The authors of paper [74] optimized pretreatment of oat husks with diluted sulfuric acid by using of Response Surface Methodology (RSM). The optimal acid treatment condition values were 130 °C, solid-to-liquid ratio (S:L) of 1:8 (w/v), concentration of H<sub>2</sub>SO<sub>4</sub> 1% (v/v) and pretreatment time 19 min. However, the authors did not discuss the content of residual lignin in the product. In another study, Soleimani and Tabil [76] showed that the optimal conditions of oat husk pretreatment were 130 °C, S:L of 10% (w/v), time of delignification 40 min and 0.05 M of H<sub>2</sub>SO<sub>4</sub>.

Budaeva et al. systematically investigated the oat husk acidic delignification in the presence of nitric acid solutions (nitric method) [11,39,78,82–90]. Temperature, reaction time and concentration of acid were varied in the range of 90–96 °C, 2–4 h and 2–6%, respectively. Optimum conditions revealed were S:L of 1:20 (w/v) at 94–96 °C, 4 h. This method allows one to reduce the lignin and hemicellulose content in husk by the factors of 1.5–2.4 and 4–5, respectively.

It should be emphasized that an application of pure acidic treatment does usually not let one to carry out the oat husk processing sufficiently for any materials to be produced. To increase a delignification degree, it is proposed to combine acidic treatment with other methods. One of the most common ways is a combination of acidic and alkali techniques. The biomass processing technique called nitric-alkaline is carried out via two subsequent stages (nitric acid first, alkali second) [44,86,91] and favors decreasing residual lignin and hemicelluloses [87,92]. According to the proposed process scheme, lignin undergoes nitration but remains insoluble. Modified biopolymer is extracted by diluted solution of any alkali at a temperatures less than 100 °C. Thus, in the patent [79] is shown that sequential two-stage treatment of oat husks with diluted nitric acid (2% (w/w), S:L of 1:15 (w/v), 90–95 °C, 6–12 h) and sodium hydroxide solution (2% (w/w), S:L of 1:10 (w/v), 80–95 °C, 5–6 h) enables to produce cellulose sample with residual lignin of 0.6–2.5%. The authors of [44,93] decreased an amount of residual lignin to 0.3% in cellulose derived from oat husks. Interestingly, the reaction product contained 93.3–94.9% of  $\alpha$ -cellulose. Polymerization degree was 1050–1420. The cellulose sample was fibers, the size of which was around 30–50  $\mu$ m in width and 25–850  $\mu$ m in length.

Sulfuric acid may substitute nitric one in two-stage acid-base oat husk processing. Thus, Soleimani and Tabil [94] spent oat husk in H<sub>2</sub>SO<sub>4</sub> solution followed by treatment in NaOH. Therein, the hemicellulose and lignin contents were reduced to 0% and 7%, respectively, cellulose extracted represented fibres. In the manuscript [95] two-stage acid-base oat husk delignification included bleaching by 1.4% acidified sodium chlorite solution, with pH adjusted to 3.0–4.0 by glacial acetic acid (70 °C, 5 h) to remove lignin. The bleached fibers were washed several times in distilled water and treated with a 5 wt.% potassium hydroxide aqueous solution (24 h, room temperature, and then at 90 °C for 2 h) to leach hemicelluloses. Cellulose yield was 34%. The purity of the cellulose sample was confirmed by IR spectroscopy. The bands at 1247 cm<sup>-1</sup>, 1516 cm<sup>-1</sup> and 1729 cm<sup>-1</sup> which correspond to

vibrations of the C–O aryl group, the C=C bond in the aromatic ring of lignin, and C=O of carbonyl and acetyl groups in the xylan were absent in the spectra. The particle size and crystallinity index of the resulting cellulose were 10-45  $\mu\text{m}$  and 52.7%, respectively.

The authors of [40,42,96,97] proposed to isolate cellulose of oat husks via treatment with peracetic acid generated in-situ. Delignification was carried out in the presence of 50% acetic acid, 38% hydrogen peroxide at 60 °C, S:L of 1:12.5 (w/v). Reaction time was 24 h. The resulting product yield was 30-33%. Amount of residual lignin was 3%. The removal of non-cellulose components was confirmed by IR and NMR spectroscopy. CI of the product of 35% was higher compared to one of pristine oat husk due to deletion of amorphous domains of cellulose in biomass. A microscopy of the product confirms the formation of long separate fibers of the polysaccharide. Cellulose was of I type.

To increase the purity of the cellulose some authors propose a multi-step treatment of oat husks. The technique includes subsequent application of acids, bases and/or hydrogen peroxide. The treatment of intermediate cellulose by bases and/or  $\text{H}_2\text{O}_2$  can be carried out several times to improve the product quality.

Kashcheyeva et al. [39] separated cellulose from oat husks by multi-step process which consisted of preliminary hydrolysis of the substrate ( $[\text{HNO}_3]$  – 0.2-0.4 wt.%, 90-95 °C, 1 h, S:L(w/v) = 1:15), a treatment by 3-6 wt.% solution of  $\text{HNO}_3$  (90-95 °C, 10-12 h, S:L(w/v) = 1:15), subsequent bleaching by 3-6 wt.% and 0.5-1.0 wt.%  $\text{NaOH}$  (90-95 °C, 2-4 h, S:L(w/v) = 1:15 and 90-95 °C, 1-2 h, S:L(w/v) = 1:10), and, finally, souring with 0.5-1.0 wt.%  $\text{HNO}_3$  (40-60 °C, 15-30 min, S:L(w/v) = 1:15). The yield of cellulose produced by such a multi-step experiment reached 23% and 1 and 2.5 wt.% of lignin and hemicelluloses were revealed in the product. Another approach applied by the authors differed from the previous one by sequence of stages. After hydrolysis ( $[\text{HNO}_3]$  – 0.2-0.4 wt.%, 90-95 °C, 1 h, S:L(w/v) = 1:15), treatment by 3-6 wt.%  $\text{NaOH}$  (90-95 °C, 6-8 h, S:L(w/v) = 1:15) preceded the reaction with a 3-6 wt%  $\text{HNO}_3$  (90-95 °C, 4-6 h, S:L(w/v) = 1:10). While the inversion of the process steps increased cellulose yield to 34%, the amounts of residual lignin and hemicelluloses achieved 2.1 and 6.9 wt.%, respectively.  $\alpha$ -cellulose content was 92.5% in both cases.

There are some other results reported which describes multi-step approach for isolation of cellulose of oat husks. The authors of [80,83] investigated delignification of oat husks as follows: 1) preliminary hydrolysis by 0.1 % dilute  $\text{HNO}_3$  (91-93 °C, 1h), 2) treatment of the pulp in solutions of 4% (w/w)  $\text{NaOH}$  and 4% (w/w) nitric acid under 92 °C.

The obtained sample contained 1.7% of lignin. In order to remove residual lignin the produced wet pulp was additionally pretreated with 2% (w/w) sodium hydroxide solution at 60 °C and washed with 0.2% (w/w) sodium hydroxide solution and water. After this procedure the sample was followed by cleaning with a water-ethanol mixture. The final cellulose contained less than 0.5% of residual lignin. Cellulose consisted of coiled plane fibers and had an advanced surface (thickness of 2–5  $\mu\text{m}$ , wideness of 20–30  $\mu\text{m}$  and a length of 300–800  $\mu\text{m}$ ). The authors believed that an application of ethanol predicted resorption of lignin solved on a cellulose surface.

Sjöstedt carried out multi-stage acid delignification of oat husk followed by alkali or peroxide treatment [73]. The first stage was a prehydrolysis of biomass under following conditions: 0.1%  $\text{HCl}$  or 0.0137-0.0274 M  $\text{H}_2\text{SO}_4$ , 160 °C, S:L of 6:1, 15 rpm, 0.5-1h. In the second stage, alkali delignification (soda pulping) was carried out at 170 °C, 4%  $\text{NaOH}$ , S:L of 8.3:1, 4h. Finally, the third stage was a two-step bleaching of obtained pulp via treatment with hydrogen peroxide with the addition of EDTA and peroxide treatment with the addition of sodium hydroxide. After alkaline delignification pulp contains 87.7 - 98.1 % cellulose and 0.1 - 0.05 % lignin. Pretreatment with sulfuric acid results in better removal of lignin. However, the degree of cellulose polymerization was 238-279, while after hydrochloric acid treatment the degree of polymerization varied from 405 to 437. According to the data of solid-state NMR analysis, the obtained cellulose samples belong to the Cellulose I type (a peak was observed in the region of 64.9ppm). Mass average molar mass of pulp was 89431, which is 4.8 times lower than in the feedstock.

Shahi et al. [98] treated of black oat as following: 1) neutral detergent solution and acid detergent solution (100 °C, 1 h) using a fiber analyze, 2) alpha-amylase, and sodium sulfite, 3) alkaline hydrogen peroxide (1 N  $\text{NaOH}$ , 3% hydrogen peroxide). In the sample obtained cellulose contain lignin was

less 2%. Cellulose derived from black oat exhibited of high thermal decomposition temperature (~370 °C), which was even higher than commercial cellulose.

### 3.1.2. Alkali treatment

According to literature data, the possibility of separation cellulose by alkali delignification is also discussed [39,48,59,73,99–102]. The advantages of alkaline treatment are the low cost of the reagent, low process temperatures, atmospheric pressure, the possibility of reusing the residual alkaline solution, and the less aggressive effect of the reagent on equipment compared to acid treatment.

The idea of alkaline delignification is to remove lignin from the lignocellulosic matrix of a plant. The mechanism of alkaline hydrolysis is that the uronic ester bond that functions as cross-link is broken by saponification, and the crosslinks between the xylan chain and other polymer units can be broken. Finally, lignin is removed and cellulose and hemicellulose are retained [103]. The destruction of the bonds between lignin and hemicellulose in lignin-carbohydrate complexes also leads to an increase in the hydrophilicity of lignin and promotes its dissolution [104,105]. Alkali treatment influences cellulose via decreasing polymerization degree of the polysaccharide. It may one to obtain cellulose being suitable for chemical transformations into bioethanol, for example. Also the cellulose derived via technique under consideration can be processed to furfural derivatives by catalysis.

In work [59] alkali pretreatment was carried out by dilute solution NaOH at 94–96 °C during 4h. This treatment leads to an increase in the cellulose content of the obtained product compared to the feedstock and to a decrease in the amount of lignin from 18.1% to 5.4%. Nowadays, scale production of alkali-treated cellulose of oat husks is also under consideration. Skiba et al. carried out pretreatment of oat hulls with 2% (w/w) alkaline solution at 80 °C during 1 h and at 2100 rpm in pilot installation comprised of a rotary-pulsating apparatus and a 100 L stainless steel reactor [102]. Loading of oat hulls were 1.5 kg. The obtained pulp was washed with 0.5% (by weight) sodium hydroxide solution, then with water. In the pulp from the oat hulls reducing the content of pentosans by a factor of 5, acid-insoluble lignin by a factor of 3.4 were observed, whereas the cellulose content increased by 1.9 times. Kashcheyeva et al. [39] increased scale of oat husk treatment in a pilot installation (S:L (w/v) =1:15, 10 kg of substrate, [NaOH] – 3-6 wt.%, 90–95 °C, 6–8 h). After the delignification, 10-13 wt.% of lignin и 6.1-7.7 wt.% of hemicelluloses were found in the cellulose product, its yield was 41%.

Similar to acidic approach, for the total removal of non-cellulose components of oat husks, additional treatment is required. Acids, hydrogen peroxide and bases are used for bleaching the cellulose semi product. Purification can be repeated several times for increasing quality of the target material.

Previously, cellulose separated from the oat hulls by alkaline delignification method with sodium hydroxide was shown to be additionally treated by nitric acid. Cellulose has a degree of polymerization of 1139 and an index of crystallinity equal to 68% [106]. This method showed a high degree of reproducibility [87,107].

Valdebenito et al. [49] proposed tree-step way of oat husk transformation. It consisted of the following subsequent treatments 1) 0.1 M NaOH, 30 °C, 18 h, 2) 0,1 M HNO<sub>3</sub>, 85 °C, 1 h, 3) 3% H<sub>2</sub>O<sub>2</sub> at 70 °C, 1 h. Residual content of lignin and hemicelluloses were less 1 and 26%, respectively, cellulose yield achieved 63%.

Oliveira et al. [48] obtained cellulose fibers from oat husks by alkali pretreatment with 4% (w/v) NaOH solution at 80 °C, 4 h. Cellulose semi product was subsequently bleached by the mixture of NaOH (0.017 g/L), acetic acid (0.075 g/L) and sodium chlorite (0.017 g/L). S:L ratio was 1:1, 90 °C. Reaction time was 4 h. Obtained sample of cellulose fibers contain 94% cellulose, 1.4% hemicelluloses and 1% lignin. The cellulose fibers had a structure similar to commercial cellulose fiber samples. The index of crystallinity was 63.5% (cellulose I type). During the transformation, from 100 g of the oat husks the researchers obtained 18.4 g of cellulose.

Along with acid and alkali treatments some alternative agents are also investigated for oat husk transformation to cellulose. Dehkhoda et al. [108] studied oat husk delignification by ammonium

persulfate (0.15 M  $(\text{NH}_4)_2\text{S}_2\text{O}_8$ , 900 rpm, 60-70 °C, 20 h). As reported by the authors, the advantage of this method of cellulose isolation is the simplicity of the method, the one-pot organization of the process. The technique does not require additional processing, including energy-consuming mechanical methods. The removal of non-cellulose components occurs with high efficiency. Such an extraction let one to derive nanocellulose which has high degree of crystallinity [109–111]. In the process of heating a solution of ammonium persulfate, the formation of free radicals and particles,  $\text{SO}_4^-$  and  $\text{H}_2\text{O}_2$ , occurs. The active species penetrate into the amorphous regions of cellulose, destroy non-crystalline domains of the polysaccharide and lignin. The process let one to form nanocrystalline cellulose. Using XRD, the resulting nanocellulose was shown to be of I type, the crystal lattice was monoclinic. It was also confirmed that the cellulose extraction method used did not lead to the transformation of the cellulose I into cellulose II. The CI of the prepared sample estimated was 57%. The average size of the resulting nanocellulose particles was 30 nm. The absence of lignin impurities in the nanocellulose sample was confirmed by FT-IR.

### 3.1.3. Combination of chemical and physical treatment approaches

In case of cellulose isolation of oat husks, interesting results can be obtained via combination of chemical and physical treatment techniques. For the processing oat husks ultrasonication, steam explosion, microwave heating are discussing.

Winuprasith et al. [112] recovered cellulose from oat bran to compare 1) chemical treatment carried out by 5% w/w solution of KOH (90 °C, 2 h) and 1 %w/w HCl (80 °C, 2h), 2) microwave treatment (800 W, 5 min, 5 cycles) and 3) hydrothermal treatment (120 °C, 2h). All the recovery techniques were followed by bleaching with hydrogen peroxide (30% v/v, 90 °C, 3 h). Unfortunately, the authors of [112] did not discuss the yield, CI and purity of the target product. Lawford et al. pretreated the feedstock by a combination of steam explosion (1 min) and 0.5-1% (w/w) sulfuric acid treatment [75]. This procedure destroyed the fiber structure of the raw material. Budaeva et al. used hydro-thermobaric processing of the substrate in a high-pressure reactor [81,113,114] under 1-2 MPa. Time of exposure varied from 0 to 1200 s and temperature was chosen in the range of 179-230 °C. After the pretreatment, the resulting fibrous product was additionally treated with 2% (w/w) sodium hydroxide solution under 55 °C during 30 min.  $\text{H}_2\text{O}_2$  was added into the suspension. Mass fraction of lignin in obtained cellulose were varied from 0.8 to 13.5%.

Machado et al. [100] compared several approaches to oat husk delignification. The authors investigated the following agents:

- alkali pretreatment with sodium hydroxide solutions (0.5%, 1%, 2%, 3% or 4% (w/v)) spent at S:L of 1:10 (w/v), 121 °C, 0.5 h;
- alkali pretreatment with  $\text{H}_2\text{O}_2$  solution (7.5% (v/v), pH = 11.5, S:L of 1:25 (w/v), 25 °C, 1 h);
- alkali treatment using ultrasonication (1% (w/v) NaOH solution, S:L of 1:7 (w/v), power 90 W, frequency 20 kHz, 20 min);
- sequential alkaline (1% NaOH solution (w/v), S:L of 1:10 (w/v), 121 °C, 0.5 h) and ultrasonic pretreatment (water, S:L of 1:7 (w/v), power 90 W, frequency 20 kHz, 20 min).

It is shown that an increase in the concentration of an alkali solution leads to a decrease in the content of lignin in a cellulose-containing product from 13.5 to 2.2%. Pretreatment of oat husk with  $\text{H}_2\text{O}_2$  reduces the residual lignin content to 7.3% in the pulp product. Single ultrasonic pretreatment demonstrated low efficiency in terms of delignification. Only 1.3% of lignin was removed. At the same time sequential alkali and ultrasonic treatment of the biomass showed the results similar to NaOH treatment only. Thus, the authors demonstrated that ultrasonication seems to be ineffective for the production of cellulose from oat husks.

An influence of a combination of chemical acid or alkali treatments and steam treatments on the delignification of oat husks was also reported in [115]. The substrate was pretreated in a steam in an autoclave (1.5 atm, 121 °C, 0.5 h, S:L of 1:9.1 (w/v)) or undergoes ultrasonication (80 °C water, S:L of 1:9.1 (w/v), 30 min). Then, biomass was dispersed in 1% sulfuric acid solution or in 10% sodium hydroxide solution or water. The content of residual lignin in the samples of the cellulose product ranged from 12.6 to 7.2%. The index of cellulose crystallinity was in the range of 47-77%. Analysis of

the obtained data showed that steam treatment is more effective compared to ultrasonic. The lowest content of residual lignin and highest crystallinity index was in the sample subjected to steam treatment combined with H<sub>2</sub>SO<sub>4</sub>.

The authors of [50,71,116] applied reactive extrusion combined with alkali-peroxide treatment of oat husks [117]. To carry out the reaction, a sample of oat husk was placed in a sealed container with sodium hydroxide solution (pH = 11.5), the moisture content of the husk was 32%, and kept for 24 h at 7-10 °C. Then a solution of hydrogen peroxide (7 g H<sub>2</sub>O<sub>2</sub> per 100 g of hulls) was added to the resulting mixture. The material was extruded in a single-screw extruder with four heating zones, where the temperature was maintained at 110 °C in all zones. After extrusion, the samples were dried (60 °C, 24 h) and crushed. It was shown that the removal of lignin resulted in the production of cellulose samples with a more open morphology and greater surface area and porosity. Reactive extrusion may be an alternative fiber modification method that has several advantages, such as short processing time and no wastewater generation. However, the authors did not discuss the residual lignin amount.

Reactive extrusion can be also combined with acid-alkali treatment. Debiagi et al. proposed two ways of the biomass delignification: 1) three-step reactive extrusion (firstly NaOH (10 wt.%), secondly double treatment with H<sub>2</sub>SO<sub>4</sub> (2 wt.%) combined with ultrasonication (15 min, 20 kHz), 2) subsequent pretreatment by peracetic acid (50% acetic acid, 38% hydrogen peroxide and 12% distilled water at 60 °C for 48 h, S:L of 1:16.6 (w/v)), extrusion with H<sub>2</sub>SO<sub>4</sub> (2 wt.%) and ultrasound (15 min 20 kHz) to produce cellulose I type [53]. It was found that the first three-step approach let one to produce cellulose which contains 17% of hemicelluloses and 9.5% of lignin. CI was 68%. Pretreatment by peracetic acid followed by activation with H<sub>2</sub>SO<sub>4</sub> and ultrasonication reduces hemicelluloses and lignin to 3 and 7.7%. CI of the target material was 80%.

In conclusion, the following main points may be emphasized. One-step acid or alkaline treatments of oat husks at low temperatures allow one to reduce the lignin content in the composition of the cellulose product to 5-13% only. The advantage of this process is the use of low temperatures (usually up to 100 °C). However, due to the presence of lignin impurities, the use of such cellulose without additional purification is limited.

The combination of acid and alkaline treatment results in a reduction in residual non-cellulosic components in the final product. Such cellulose can be used for the synthesis of ethanol or for the production of composites based on it. Reiteration of treatment (acid-base and/or peroxide-base) combined with additional bleaching by bases or hydrogen peroxide propose a way for obtaining cellulose which has high purity and suitable for the synthesis of nanocellulose, including bacterial cellulose, composites and hydrogels, cellulose esters. The degree of polymerization and crystallinity index in this process vary in the ranges of 800-1400 and 60-70%, respectively. Disadvantage of the method is a quite low yield of the target polysaccharide. It may vary from 18 to 35%. Purified cellulose is also suitable for catalytic transformations of the polysaccharide to alcohols (sorbitol, ethylene and propylene glycols), organic acids (formic, levulinic, glycolic) and furan derivatives

A combination of chemical and physical (steam explosion, hydro-thermobaric processing, ultrasonic pretreatment, microwave treatment, reactive extrusion) processes is also under consideration. An analysis of the literature data showed that the most promising method that can be implemented on an industrial scale is the use of a combination of chemical processing and reactive extrusion. Steam treatment is also perspective. This method is most actively used by research groups in Brazil. The advantages of the method include simplicity of the organization, small reaction time, low quantities of solvents involved.

### 3.2. *Organosolv treatment*

Organosolv treatment is widely discussed for the transformation of plant materials to cellulose [60,103,118–120]. The method of organosolv treatment has already been applied for processing of miscanthus [121], wheat straw [122–124], eucalyptus biomass [125], bamboo [126], pine [127,128], bagasse [129], rice straw [130,131], birch [132], abies wood [133]. The method has low cost and the

ability to reuse solvents [134]. However, the process is carried out at high temperatures, and the equipment used in the process have to be resistant to elevated pressure.

The authors of [13] provides a technical and economic assessment of the possibility of using oat husk organosolv treatment to extract cellulose for bioethanol production. A profit could be achieved in 16.8 years after implementation of the method.

The organosolv process proceeds as follows. Plant raw materials are loaded into the reactor filled with a mixture of water and an organic solvent (ethanol, methanol, glycerol, etc.). The reaction media is acidified with an inorganic acid or a solid acid catalyst is added. The reactor is heated and maintained for the required time. Cellulose is obtained as a solid product. Lignin which is dissolved in a water-organic solvent is removed from the solution by diluting it in water or a solution of lye and filtered. A solution of hemicelluloses remains in the liquid fraction. Organosolv lignin is a product with increased added value. Such lignin has a molecular weight of 700-1550 g/mol, low polydispersity, a glass transition temperature of 70-170 °C, a high relative amount of phenolic hydroxyl groups and a low degree of chemical modification [14,135,136]. This lignin can be used in the production of molding compounds, urethane epoxy and formaldehyde resins, and antioxidants.

Nowadays, organosolv process is not widely applied for the transformation of oat husks. Chopda et al. [7] studied recovering cellulose, lignin and hemicelluloses of oat husks via organosolv treatment in water/ethanol mixture with H<sub>2</sub>SO<sub>4</sub>. The authors proposed optimum conditions of treatment: 210 °C, 90 min, and S:L ratio of 1:2. The reaction was carried out at low acid loading: 0.014 g of H<sub>2</sub>SO<sub>4</sub> per 100 g of oat husk. Two fractions were obtained, first one of lignin (main product content 74,5%) and the second one of polysaccharids (74,9% purity). Sar et al [134] shown the possibility of using phosphoric and oxalic acid as an alternative to sulfuric acid in the organosolv pretreatment of oat husks. Authors demonstrated that these acids are effective for obtaining high quality lignin and glucan. The optimum conditions for treatment of the husk were the following: 1/1 (v/v) mixture of water and ethanol acidified with oxalic acid, 210 °C, reaction time 90 min, S<sub>i</sub>L ratio of 1:2.

Thus, to our knowledge, only a few works are devoted to organosolv cooking of oat husks. As noted above, the advantage of this process is that during organosolv processing three products can be obtained, namely lignin, hemicellulose, cellulose. However additional investigations are required to increase the purity of the target products, for example via combination of organosolv treatment with alkali or hydrogen peroxide bleaching.

### 3.3. *Hydrotropic method*

The hydrotropic method of cellulose isolation from biomass occurs a moderate adjustment on cellulose and lignin during the reaction due to neutral pH. It allows one to isolate cellulose and lignin without significant modifying their structure. This process is carried out using non-toxic and low cost reagents, at low pressures; chemicals can be reused [34,35,55,61,137–141]. Denisova et al carried out cellulose separation from oat husks by several stages [34,35,137,138,141]. Firstly, pre-hydrolysis at 140-160 °C with solid-to-liquid ratio of 1:8 was performed. At the second stage, the pre-hydrolyzed biomass was treated with 30% sodium benzoate solution at 160-180 °C for 1 and 3 h. In this stage technical cellulose was obtained with a yield of 59-77% (w/w). The product contained 14-15% lignin and trace amounts of hemicelluloses. It should be noted that removal of hemicelluloses was more complete after carrying out hydrotropic treatment for 3 h. In the third stage, cellulose product (technical cellulose) was additionally treated with H<sub>2</sub>O<sub>2</sub> in 1% (w/w) sodium hydroxide solution, then with HCl solution and washed with ethanol. It removes a significant amount of lignin, ash and hemicelluloses. After hydrotropic treatment the obtained pulp samples should be washed with either hydrotropic reagent or dilute sodium hydroxide solution in order to avoid lignin deposition on the cellulose fibers [54]. Such a treatment allows to obtain cellulose with yield of 40-45 %, in which the mass fraction of the residual lignin is 5-9 %. In the obtained product the content of cellulose was 79-90%. The degree of polymerization and index of crystallinity of the separated cellulose were 890-1640 and 79%, respectively [141]. Temperature, sodium benzoate concentration and pretreatment time were determined as the main factors influencing the process [61].

Interestingly, the hydrotropic treatment demonstrates appropriate results for the isolation of cellulose from oat straw [61,142]. The process can be conducted under following conditions: temperature – 160 - 180 °C, S:L=1:10, time of the reaction – 1 or 3 h. Technical cellulose samples were isolated from the substrates, which contained  $\alpha$ -cellulose and residual lignin with yields of 60.6-79.2 and 8.3-13.1, respectively. IR spectroscopy confirmed the removal of a significant amount of non-cellulosic components during hydrotropic treatment of oat straw [143]. The crystallinity index of the studied cellulose samples was estimated by IR spectra. Value of crystallinity index is 0.71. In the work [61] the process of the hydrotropic delignification of oat straw in the universal thermobaric unit was investigated.

Thus, only one group of researchers applied hydrotropic treatment to the oat husk transformation. The results demonstrated that single hydrotropic processing produces cellulose which demonstrated low purity. Significant additional efforts in terms of bleaching is required.

#### 4. Materials based on cellulose isolated of oat husks

As mentioned above, there are plenty of materials based on cellulose derived from biomass. Cellulose ethers [144] including nitrocellulose [145,146], composites [147–150], nanocellulose [151–153], hydrogels and aerogels [154–156] are proposed.

Interesting in the synthesis of materials based on cellulose is associated with 1) the possibility of obtaining materials which have advanced target properties, and 2) the production of biodegradable products [148]. This chapter will describe some prospects in the synthesis of materials based on cellulose isolated from oat husk.

##### 4.1. Cellulose ethers

Cellulose ethers are widely applied for the production of fibers, plastics, coatings [157]. Thus, for the production of coatings cellulose acetate (CA), acetopropionate (CAP), acetobutyrate (CAB) and nitrocellulose are applied [1]. CAB are used in paint industry. The structural, physical and chemical characteristics of cellulose ethers are controlled by modifying the polymer structure, for example by changing the degree of substitution and chain length. By etherification, it is possible to change the nature of cellulose from hydrophilic to hydrophobic by introducing hydrophobic additional groups. For example, when increasing butyryl content in CAB, flexibility, hydrocarbon resistance, solubility increase, but chemical resistance and hardness decrease. Increasing the molecular weight of CAB leads to a rising down in solubility, but an increase in melting point and durability.

Nowadays, the main raw material for the production of cellulose esters is woody biomass. However, it seems that non-wood agricultural wastes raw materials are able to replace wood biomass for the bioethers production. This section describes the possibility of the production cellulose ethers from oat husk.

Protopopov, Bobrovskaya et al. [43,158] produced ethers of cellulose in pre-hydrolyzed oat husks with *m*-hydroxybenzoic and *o*-hydroxybenzoic acids. The substitution degree in the polymer varies from 1.5 to 2.5. The materials under study seems to be perspective in construction, energetic, for the production of plastics and elastomer-like materials. Acetylation reaction time was 1-1.5 h, and the temperature varied in the range of 25- 55 °C. Increasing temperature and process time was shown to favor substrate conversion. Ether formation was confirmed by IR-spectroscopy.

Klevtsova et al. [159] produced ethers of pre-hydrolyzed oat husks with benzoic and sulfosalicylic acids. The experiments were carried out at 55 °C, 2-5 h. The authors revealed an amount of residual lignin in the target product was 5-9%. Polymerization degree of the cellulose ethers varied from 200 to 1400 and increased simultaneously to decreasing temperature and treatment time. The degree of substitution in the resulting cellulose esters ranged from 0.44 to 9.63% with sulfosalicylic acid and from 0.29 to 73% with benzoic acid.

Giraldo et al. used citric acid to modify cellulose isolated of oat husks. The influence of ultrasonication on the reaction was also investigated [42]. Citric acid is an inexpensive and stable etherifying agent that increases hydrophobic properties of the material [160]. The authors also proposed that since the supramolecular structure of cellulose contains many hydrogen bonds, using

treatment by physical methods, such as ultrasonic may improve an efficient of the etherification. Ethers produced under ultrasonication had lower water absorption capacity of 1,63–1,71 g/g compared to pristine cellulose. At the same time, the oil retention capacity increased from 1,80 g/g (cellulose before the treatment) to 4,57-7,31 g/g after etherification.

Pareira et al. [41,96] investigated etherification of oat husk cellulose by citric and succinic acids. In the work, the authors varied the loading of the esterifying agent (0, 5, 12.5 and 20.0% g acid/100 g cellulose). The acid-wet cellulose kept for an hour was passed through a single-screw extruder (100 °C, 60 rpm). It was shown that the degree of substitution in cellulose ethers increased with increasing acid concentration and did not depend on the type of acid used. The degree of substitution varied from 2.28 to 3. In the IR spectra for all modified samples, a band was observed between 1720 and 1737  $\text{cm}^{-1}$ , which the authors attribute to the stretching of C=O carbonyl in ester bonds. The process of cellulose etherification and reactive extrusion has little effect on the CI of the modified samples. Crystallinity index decreases from 42 to 36-39%. It is worth noting that the treatment did not affect the polymorphic type of cellulose and the morphology of cellulose fibers. Modified samples showed an affinity for dichloromethane and chloroform, which are non-polar solvents, indicating that the hydrophobic character of cellulose changed by chemical modification through reaction with citric and succinic acids. Similar to [42] value of water absorption capacity for the samples of cellulose ethers (6.46-7.49 g/g) was lower compared to non-modified polysaccharide (9.27 g/g). Modification of cellulose leads to an increase in oil absorption capacity by 4-5.5 times compared to the original cellulose. This increase in the hydrophobic capacity of the modified samples can be explained by a decrease in the number of free hydroxyl groups on the cellulose surface upon reaction with organic acids.

#### 4.2. Nitrocellulose

Nitrocellulose (CN) is an important kind of cellulose based materials [59,161–167]. They have advanced strength properties and can be converted into a plasticized state, they are highly soluble and compatible with available plasticizers. Depending on the degree of substitution, cellulose nitrates can be used as the energy basis of nitrocellulose powders, rocket fuel components, for the production of optical transparent films, biological indicators, semi-permeable membranes, selective sorbents, and also as a component of varnishes, paints, nitro enamels and other products [168–170]. Membranes made of nitrocellulose are widely applied for the production of biosensors due to high efficiency of protein binding [171]. One of the current trends is the development of nanothermites using CN that have low sensitivity to mechanical stress [172].

The traditional starting raw materials for the production of various types of CN are cotton and wood natural fibrous materials [93,173]. It has been previously shown that nitrocellulose can be obtained from non-wood raw materials, such as flax [44,174], tobacco waste [175], industrial hemp [176], wheat straw [177], esparto [178,179], miscanthus [92,93,173,180–182]. Interestingly, oat husk is also a potential substrate for manufacturing CN [44,183]. According to the literature, for high-quality CN to be produced, starting material should contain more than 92% of  $\alpha$ -cellulose and non-cellulosic component should be less than 1.2% [181]. In addition, the nitration process is affected not only by the purity of the cellulose, but by its shape and structure, since the more developed and accessible the cellulose surface is, the more uniform the nitration process occurs. A significant number of works on the production of nitrocellulose from non-wood raw materials, including oat husks, were published by a group of scientists led by academician G.V. Sakovich [45–47,165,166,183,184]. The authors carried out the synthesis of nitrocellulose via treatment of the pulp by sulfuric-nitric acid mixture at 20-40 °C, 30-90 min, mass ratio 1:25 - 1:40 [46]. The pulp was prepared by two-step acid-base technique.

Since the chemical stability of CN is an important characteristic, the resulting nitrocellulose samples were subjected to additional processing. Stabilization was carried out in three stages: treatment in water at a temperature of 90-95 °C for 1 hour, high-temperature treatment in an autoclave in a 0.3% solution of nitric acid at a temperature of 130 °C and in a 0.03% solution of sodium carbonate at a temperature of 90-95 °C for 3 hours. The influence of the treatment time of the nitric

acid solution on the characteristics of the resulting nitrocellulose was studied in [46,59]. Thus, carrying out the process for 140-160 minutes makes it possible to achieve the characteristics similar to ones of Colloxiline-N commercial product. The following properties such as the viscosity of a 2% solution in acetone, solubility in an alcohol-ether mixture, and the volume of NO were investigated. The resulting NC retains the shape of the original cellulose fibers; the width of the fibers is 22-82  $\mu\text{m}$ , and the length is 382-522  $\mu\text{m}$ . The nitrogen content in the CN samples was 12%, the viscosity of a 2% solution in acetone was 10-13 mPa·s, and the solubility in the alcohol-ether mixture was 98%. IR spectroscopy confirmed the formation of cellulose nitrate esters during the synthesis of CN (bands at 1650-1635, 1279-1276, 831-825, 746, 687-672  $\text{cm}^{-1}$ ). CI of the cellulose was 64%. Monoclinic phase structure of the CN was changed to pseudo-orthorhombic one [169]. The temperature of the thermal destruction of CN was 200 °C. Obtained cellulose nitrates from oat husk have high specific heats of decomposition of 6.28-7.54 kJ/g (for colloxylin, this parameter is within 7.64-7.73 kJ/g [59,185]).

#### Bacterial nitrocellulose (BCN)

Bacterial cellulose is of particular interest for the production of nitrocellulose due to the unique properties [171,186]. More detailed information on the properties of bacterial cellulose is discussed in the section 4.4.1. BCN material is a perspective component of fuels as it features an ultrafine, high-purity fibrous network and a more stable structure compared with plant-based cellulose nitrates [187].

Due to the high tensile strength, ultra-fine structure of microfibers, as well as their ultra-purity, BCN will provide better mechanical and performance characteristics of the fuel compared to traditional energy binders. In addition, ionizing radiation detectors, biosensors, chips, membranes, etc. are made based on BCN [188,189]. Yamamoto et al. demonstrated perspectives of the synthesis of nitrates of bacterial cellulose cultivated by *Acetobacter xylinum* in Hestrin-Schramm's medium [190]. In the papers [191-194], an influence of reaction conditions such as ratio of acids, temperature and time of the process, ratio of substrate and nitrating agent on degree of substitution of the polysaccharide was investigated. Gismatulina compared two techniques of nitrating bacterial cellulose. The substrate of bacterial cellulose was produced by a *Medusomyces gisevii* Sa-12 symbiotic culture. The first nitration approach was a use of a mixture of sulfuric and nitric acids, the second one was a nitration using concentrated nitric acid and methylene chloride [191]. Both BCN samples exhibit a high viscosity value compared to nitrocellulose obtained from plant materials. However, BCN sample derived via the second method had both better viscosity and nitrogen content (12.27%) compared to the material derived via traditional approach. The authors proposed for the high N-content to be associated with an advanced reaction ability of the bacterial cellulose with methylene chloride, since such treatment facilitates the penetration of the nitrating agent into the original cellulose. On the other hand, advanced velocity is caused by hydrolytic degradation when nitrating and its mild stabilization. However, to our knowledge, no publications have been found on the production of BCN using hydrolyzate from oat husks what provides a perspective field of research.

#### 4.3. Cellulose biodegradable composites

Oat hulls can serve as a source of natural cellulose fibers which can be used as fillers or reinforcing elements in a polymer matrix for the production of biocomposites. Some authors assume such advantages of cellulose fibers for biocomposite manufacturing as low density and cost, a large-scale availability, biodegradable, abrasion resistant, high stiffness and high filling potential. Thanks to these advantages, natural fiber composites could be used in the production of plastics, packaging for medicine and the automotive and construction industries [116,157,195,196]. Prior any spending, oat husks should be delignified and reactive extrusion seems to be a perspective environmentally friendly way for the production of the composites [197,198].

The authors of [50] prepared samples of composites based on starch and poly(butylene adipate-co-terephthalate). Oat hulls subsequently pretreated by NaOH and H<sub>2</sub>O<sub>2</sub> were also incorporated to the material. However, the authors revealed that oat hull cellulose did not cause any improvement of mechanical and thermo stability. Similar conclusion was described in [52] for the composites of polylactic acid with a cross-linked starch/oat hull mixture synthesized by reactive extrusion with

sodium trimetaphosphate for application in composites with. Apparently, the lack of reinforcement may be due partial destroying the structure of the cellulose fiber during chemical treatment and reactive extrusion. Reddy et al. [199] produced biocomposites based on polypropylene/polylactide reinforced with oat husk fiber. The material derived showed the reduction in tensile and flexural properties when compared to the PP/PLA blend.

Giubilini et al. [51] proposed an utilization of oat husks as a component of a biodegradable composite based on poly(3-hydroxybutyrate-co-3-hydroxyhexanoate). In this work, oat husk fibers were used as a filler in the polymer matrix and provided a partial replacement for the copolymer. It was shown that the resulting composite (8 vol.% of oat hulls) did not deteriorate in thermal and mechanical properties compared to the polymer without filler. However, according to the data obtained, for such a composite the Young's modulus increases by 12% without loss of elongation at break.

It has been shown in the literature that cross-linking of cellulose improves the stability of the polymer mesh and affects physical properties such as polymer elasticity, viscosity and solubility [200,201]. Cross-linked cellulose can be used to produce biodegradable hydrogels being perspective in various biomedical applications (e.g., targeted drug delivery). Such polymer can also be used in agriculture for controlled release of agrochemicals such as pesticides, herbicides, microbicides and plant growth regulators [202]. In addition, cross-linked cellulose can be used as a component of composites for the manufacture of packaging materials [203].

Marim et al. [97] proposed to obtain cross-linked cellulose isolated from oat husk using peracetic acid. Reactive extrusion with tartaric acid or sodium trimetaphosphate was applied [96]. Sodium trimetaphosphate which has low toxicity and cost is used as a cross-linking agent. Cross-linking with trimetaphosphate occurs by forming an intra- and/or intermolecular cross-linkage of one phosphate linked between hydroxyl groups in polysaccharides molecules. Tartaric acid can act as both an esterifying agent and a cross-linking agent. Tartaric acid reacts with cellulose by attaching the carboxyl group of the acid to the hydroxyl group of cellulose (esterification). Further, the hydroxyl group of another cellulose molecule may interact with another carboxyl group of the same tartaric acid molecule, which may result in the formation of cross-links between the cellulose chains. Extruders used for the reactions combine thermomechanical and chemical processing to promote esterification of the carboxyl groups of tartaric acid and the hydroxyl groups of the cellulose chain. The use of reactive extrusion allows the modification process to be carried out continuously. In addition, no solvent is used during the modification. Different ratios of tartaric acid or sodium trimetaphosphate to cellulose were used to obtain cross-linked cellulose samples (acid/cellulose (g/100g) = 5, 12.5, 20; sodium trimetaphosphate/cellulose (g/100g) = 0.025, 0.05, and 0.1). The required suspensions of crosslinking agents were dissolved in water and mixed with cellulose so that the resulting sample had 32% moisture content. After 24 hours, the samples were extruded in a single-screw extruder (100 °C,  $v$  screw = 60 rpm). For the cellulose samples modified by tartaric acid, a band at 1735  $\text{cm}^{-1}$  was observed in the IR spectra. It can be attributed to the valence vibration of ester carbonyl groups [200,203]. No bands attributed to phosphate groups were found in the spectra of samples modified with sodium trimetaphosphate, which is probably due to the low concentration of crosslinking agent. The degrees of substitution for the cellulose ether samples were 1.58-2.96. For all samples obtained with sodium trimetaphosphate, the degree of substitution was 0.01. In micrographs, the authors observed individual fibers of the modified cellulose samples that were similar to the unmodified cellulose. Thus, extrusion of cellulose with tartaric or sodium trimetaphosphate does not affect the morphology of cellulose fibers. However, performing cellulose modification results in a slight decrease in CI c 42 to 37-32%.

The etherification process in combination with reactive extrusion treatment does not affect the polymorphic type of cellulose (does not change the crystalline organization of cellulose). Lower CI values were observed in the samples with higher crosslinking agent loading. Apparently, this method of obtaining modified cellulose samples affects the ratio of crystalline and amorphous sites, and, possibly, higher crosslinking agent loadings lead to fewer free hydroxyl groups, which are responsible for the formation of hydrogen bonds in the crystalline structure of cellulose [204]. The

modification of cellulose allows increasing the oil absorption capacity by the factor of 4.1-4.8 compared to the original cellulose [41].

Hydrophobic cellulose derivatives can be used as encapsulation materials for hydrophobic bioactive compounds [205] or agrochemicals such as pesticides or fertilizers. All modified samples showed affinity for chloroform. This also indicates that cellulose modified with tartaric acid and sodium trimetaphosphate is hydrophobic, since the replacement of hydroxyl groups in cellulose by less polar groups results in more hydrophobic materials [41,206]. Such materials can be successfully used in the synthesis of composites based on nonpolar polymeric hydrophobic matrices. Modified hydrophobic cellulose disperses better in nonpolar matrices. The obtained composites based on them are suitable for the manufacture of food packaging [116].

#### 4.4. Nanocellulose

Nanocellulose (NC) represents a special kind of cellulose material which consists of particles with a length of several microns and a diameter up to 100 nm. It has special properties such as high elasticity, tensile strength, controlled crystallinity, high degree of polymerization, large specific surface area [40,150,207–209]. Due to these physicochemical properties, nanocellulose has perspective applications (aerospace, medicine, etc.). NC is attributed as an additive to biodegradable composites [40,210,211]. Carboxylated nanocellulose is shown to be possible carrier of pharmaceuticals [108]. Depending on the physico-chemical properties, nanocellulose materials are devoted into three main groups: cellulose nanofibrils (CNF) [210], bacterial cellulose (BC) [211,212] and cellulose nanocrystals (CNC) [213,214].

##### 4.4.1. Bacterial nanocellulose

Bacterial cellulose (BC) has a composition similar to one of the polysaccharide derived from plant biomass [2,57]. BC biopolymer is represented by fibers of 3-5  $\mu\text{m}$  which, in turn, consist of nanofibrils of 20-100 nm length and 3-4 nm thickness [215]. Degree of polymerization can be up to 5500 [211]. It was shown that the degree of polymerization of BC samples depends on the dosage of inoculum and the duration of biosynthesis. Thus, it is possible to control the biosynthesis process and the degree of polymerization [69,216]. Interestingly, the degree of polymerization of plant cellulose isolated of oat husks is in the range of 1000 – 1500 [44]. BC has high crystallinity degree up to 80% and does not contain impurities of lignin and hemicelluloses [217]. BC has such perspective properties as high water-holding capacity, mechanical strength, elasticity, porosity, biocompatibility [169,218–220]. Bacterial cellulose could find applications in the production of biodegradable packages [64,221–225], regenerative medicine (biosensors, medical bandage etc.) [26,212,219,225–228]. Chemical modification of the hydroxyl groups of glucan units let one to prepare new materials [229–231]. It was shown that microbial cellulose can be used as a modifier of cellulose-based dielectrics [232]. In moreover, an involvement of a bacterial cellulose into paper production allows one to increase its strength and deformation characteristics.

Commonly, solutions of monosaccharides such as glucose, fructose and sucrose are used as a nutrient medium for the biosynthesis of bacterial cellulose. The media for microbial synthesis can be obtained by hydrolysis of polysaccharide plant raw materials including wastes of the food industry, agriculture. BC can be synthesized in nutrient media derived from rice bark [233], bagasse [234], wheat straw [235], tobacco waste [236], corn stalk [237], coffee husks [238], orange peel [239], citrus peel and pomace [240], pineapple peel [241], rotten banana / rotten mango [242], apple pomace [243], grape pomace [244], potato peel [245], cotton waste textiles [246]. BC production varies in the range of 2.4-10  $\text{g}\cdot\text{L}^{-1}$ .

For the first time bacterial nanocellulose has been synthesized from oat hulls in work [247]. The obtained cellulose has a high degree of crystallinity of  $88 \pm 5\%$  and is composed of 99%  $\text{I}\alpha$ -polymer. In another works [39,83,217,248] it was shown that bacterial nanocellulose synthesized by *Medusomyces gisevii* Sa-12 in both synthetic nutrient medium and enzymatic hydrolysates have a high degree of crystallinity (from 86 to 93%). Phase  $\text{I}\alpha$  reached 93.6-100%. Synthesis of bacterial nanocellulose is carried out via several steps. At the first stage, pulp is obtained from oat hulls by

pretreatment of the feedstock with diluted sodium hydroxide and nitric acid solutions under atmospheric pressure and temperature of 94°C. At the second stage, the pulp was hydrolyzed using Cellulux-A [80]. In the third step, nutrient medium was prepared by adding dried black tea to the hydrolysate followed by extraction at 100 °C during 1 min and further filtration. Finally, the obtained culture medium was used to carry out microbial biosynthesis of nanocellulose microfibrils by *Medusomyces gisevii* Sa-12 [63]. When cultivation completed, the bacterial nanocellulosic films are separated from the culture medium and washed with distilled water and dilute solutions of sodium hydroxide and hydrochloric acid.

The paper [39] demonstrates the possibility of carrying out the biosynthesis of BC on hydrolysates obtained from cellulose isolated from oat husks by acid, alkaline and multi-stage acid-base treatment. All isolated celluloses differed in the content of lignin. It is worth noting, that the method of preprocessing the feedstock primarily affects the amount of residual lignin in the cellulose product. However, volumetric productivity was not indicated in this work. In other studies, the authors showed that the biosynthesis of bacterial cellulose is influenced by the nutrient medium. Replacing a synthetic medium with a hydrolysate leads to a decrease in the quality of BC, which is associated with the presence of inhibitors in hydrolysates (for example lignin, furfural derivatives) [56,57]. Skiba et al. [57] demonstrated that the yield of BC decreased by the factor of 3.2 when using medium derived by hydrolysis of cellulose isolated from oat husk by acid treatment compared to synthetic nutrient mixture. The formation of lignin derivatives was mentioned as an explanation of decreasing BC productivity. To decrease amount of lignin in nutrient mixture the authors applied two-step oat husk treatment procedure: 1) by 4% (w/w) nitric acid solution and 2) 4% (w/w) NaOH. The amount of residual lignin decreased to 0.8%. The BC yield was 11% (volume productivity 2.2 g L<sup>-1</sup>), and the degree of crystallinity was 93%.

Thus, preprocessing of the initial plant raw materials plays an important role in the synthesis of bacterial cellulose [39,249]. To obtain bacterial biomass with high yield, it is necessary to use a hydrolysate which contain only a trace amount of lignin. Pretreatment methods such as single-step alkaline or acidic are apparently not suitable for using the resulting hydrolysates for the biosynthesis of bacterial cellulose in high yields. Interestingly, Skiba et al. demonstrated the possibility of obtaining bacteria cellulose from husk when the process is scaled up [57]. They show that yield of BC decreased by 1% in the pilot installation compared to laboratory conditions and reached 10 %.

#### 4.4.2. Nano fibrillated cellulose

Nano and microfibrillated cellulose (CNF) consists of fibrillar units containing both amorphous and crystalline domains of the polysaccharide. These materials demonstrate high strength, flexibility and perspective ratio of length to diameter [250]. CNF can be used as adsorbents [251,252], emulsion stabilizers [112,253,254], composite components [255,256] and also have potential biomedical applications [208,257–259].

For the synthesis of CNF the methods of cryocrushing [260], high pressure homogenization [49,261], milling [262], ultrasonication [263], reactive extrusion [117], MW [112], hydrothermal treatment [112] or a combination of chemical and mechanical treatments [40,49,53,112,264] are applied.

Paschoal et al. [40] prepared a cellulose nanofibers from oat husks by peracetic acid bleaching at 60 °C during 24 h and acidic hydrolysis (45 °C, S:L of 1:10, 30-60 min, 63.7 (w/v) H<sub>2</sub>SO<sub>4</sub>) followed by ultrasonic treatment (15 min). Pretreatment lead to a decrease in lignin amount to 1.1-3.5%. The authors obtained 32 g of nanofibrillated cellulose from 100 g of oat wastes. It was demonstrated that the formation of interconnected meshes of tiny nanofibers with diameters of 70-100 nm and lengths of a few micrometers. Thus, this method allows one to obtain nanofibers with a relatively high aspect ratio. These types of materials are suitable for the polymer reinforcement [265]. TEM micrographs of nanofibrillated cellulose demonstrate the homogeneity and nanometric dimensions of these materials. According to NMR and XRD data, crystallinity index of samples nanofibrillated cellulose are 41%.

Debiagi et al. produced CNF samples from oat husks using two reaction schemes: 1) three-stage reactive extrusion and ultrasonic processing of the feedstock, 2) a combination of peracetic acid pretreatment combined with one-stage reactive extrusion with  $\text{H}_2\text{SO}_4$  and ultrasonication [53]. According to AFM, application of the first technique let one to derive cellulose nanofibers which had a diameter of  $100 \pm 25$  nm (the product yield of 60%). In case of the second reaction the diameter of the cellulose product fibers was  $12 \pm 2$  nm and the yield of the material was 65%. Thus, this work demonstrates the effectiveness of using reactive extrusion for the production of CNF. The main advantages of the proposed methods were simplicity and decreasing wastewaters.

Valdebenito et al. carried out chemical pretreatment of oat hulls by mediated oxidation with 2,2,6,6-tetramethylpiperidiny-1-oxyl (TEMPO) and the mechanical treatment in a high-pressure homogenizer [49]. The authors produced CNF fibers which had 20 nm of diameter and length of several microns [266]. In addition, TEMPO derived CNF had a good capability to form rigid networks with advanced mechanical properties [267]. The synthesis of cellulose nanofibrils consisted of three stages. The first step was devoted to delignification of the feedstock by the reaction of oat husks with solutions of alkalis or acids and hydrogen peroxide as described in Section 3. The delignified cellulose underwent TEMPO-mediated oxidation of the pulp (5 g of pulping, 300 mL of  $\text{H}_2\text{O}$ , 0.16 g/L of 2,2,6,6-tetramethylpiperidiny-1-oxyl, 1.66 g/L of NaBr, 30 mL of 13% solution NaClO, pH – 10.5). TEMPO-mediated oxidation leads to the formation of carboxyl and aldehyde groups on the surface of the cellulosic material. It facilitates following modification CNFs [259]. On the third stage the oxidized pulp was dispersed in water and then homogenized with a high-pressure homogenizer (600 bar). The optical and surface properties were assessed on the CNF films. The light transmittance was quantified to 89.92. This quite low value measured for CNF films isolated from oat hulls indicates a relatively high amount of residual fibers in the fibrillated material [268]. The fraction of residual fibers was quantified to be  $10.58 \pm 0.08$  % in nanofibrated cellulose. Residual fibres also lead to an increase of roughness of CNF films [269]. The AFM analysis revealed the homogeneous morphology of TEMPO CNF, having diameters less than 20 nm and lengths in the micrometer scale. The authors proposed that incomplete fibrillation of the pulp seems to be associated with significant amount of hemicelluloses (26 %). В другой работе Winuprasith et al. [112] investigated an influence of chemical (solutions of potassium hydroxide and hydrochloric acid followed by bleaching with hydrogen peroxide) and physical (microwave and hydrothermal) treatments for the isolation of nanofibrillated cellulose from oat bran. Emulsion capacity and stability of the product was in the focus of the research. Cellulose isolated was defibrillated by high pressure homogenizer. Pickering emulsions consisted of 1% w/w CNF in water-oil media were prepared from nanofibrillated cellulose. Chemical pretreatment of the biomass significantly facilitated fibrillation by high pressure homogenizer. Additionally, positive effect of hydrogen peroxide pretreatment was demonstrated. Purification of the pulp let one to improve a ratio of nanocellulose to microcellulose.

The particle size distribution of all emulsions exhibited as multimodal distributions due to the CNF contained both micro and nano size fiber. An emulsion of chemically pretreated nanofibrillated cellulose had the smallest oil droplet diameter ( $d_{32}$ ) of  $3.76 \mu\text{m}$  compared to the samples produced using hydrothermal and microwave treatments. The authors proposed that a decreasing of oil droplet diameter was associated with a better distribution of the nano and micro size fibers. Another important parameter of the prepared sample was a  $\zeta$ -potential. It was revealed to be also affected by cellulose isolation techniques. A value of  $\zeta$ -potential was in the range of -37 to -54 mV. That pointed to the formation of the stable emulsions. Interestingly, the emulsions stabilized by CNF exhibited lower viscosity compared to CNF suspensions. The reason was that CNF emulsions had higher magnitudes of  $\zeta$ -potential. Therefore, the high surface charge emulsions stabilized by CNF treated by microwave with bleaching and autoclave with bleaching exhibited lower viscosity than others.

#### 4.4.3. Nanocrystal cellulose

Cellulose nanocrystals (CNC) consists of cylinder shape nanoparticles (100-250 nm length, 5-70 nm diameter) and has advanced values of transparency, purity, crystallinity, strength [270]. Nanocrystal cellulose also possesses high mechanical robustness, high specific surface area and

unique liquid crystalline properties. Nanocrystal cellulose seems to be perspective for the using in biomedicine, pharmaceuticals, electronics, production of films and membranes, nanocomposites, aerogels [271,272]. CNC is derived from natural cellulose during acid hydrolysis under high temperature. Acid treatment destroys amorphous domains of cellulose natural chain and stimulate the remaining crystalline areas to stick together [273]. Sulfuric acid is usually applied for the preparation of CNC, however HCl and H<sub>3</sub>PO<sub>4</sub> are also of particular interest [274]. Another approach for the synthesis of CNC is a fermentation [272]. In [275] a fermentative hydrolysis of cellulose derived from oat husks was proposed. The diameter of the target nanocrystals was 29 nm, crystallinity value reached 90%.

Bruni et al. obtained biocomposite films based on phosphorylated wheat starch and nanocrystal cellulose isolated from oat husks [271]. Nanocrystal cellulose from oat hulls was derived as described in [272].

Biocomposite films were obtained by casting as follows: an aqueous suspension of oat husk nanocrystals (3.6 g/L) was heated with solutions of starch and glycerol (90 °C, 30 min), the films were poured onto acrylic plates and dried in an oven (35 °C, 24 hours). The introduction of nanocrystals cellulose into the structure was found to cause an increase in tensile strength ( $5.07 \pm 0.33$  MPa) compared to the film obtained using native starch ( $2.65 \pm 0.09$  MPa). The authors pointed that the advanced mechanical properties, high stability and flexibility provided great potential for their use in the food industry as packaging material.

In another study, Shahi et al. [276] demonstrated the possibility of producing a biocomposite film (PVA-RC) based on regenerated cellulose fibers isolated from black oat and PVA (polyvinyl alcohol). It has been demonstrated that regenerated oat cellulose fibers are well dispersed in the polymer mixture and demonstrate good PVA/nanocellulose compatibility due to the formation of hydrogen bonds. The thickness of the resulting biocomposite films ranged from 110 to 130  $\mu\text{m}$ . It has been shown that the introduction of cellulose into the polymer matrix leads to a decrease in the hydrophilicity of the resulting material, and as a consequence, to a decrease in the swelling of the composite film compared to the original polyvinyl film. A study of the synthesized materials for thermal stability, assessed by thermogravimetric analysis, showed that the maximum temperature of thermal decomposition of composite films increased. In addition, the tensile strength of the biocomposite film containing 3% regenerated cellulose fibers increases by  $\sim 2$  MPa. A similar effect when introducing a cellulose component into a polymer composite was observed previously [277]. Increasing Young's modulus from 70 to 112 MPa is attributed to the addition of fibers and presence of disintegrated regenerated cellulose, creating interface hydrogen bonds with the PVA matrix, thus anchoring the PVA chains against the movement. Elongation at break of the film tended to decrease this parameter with increasing cellulose fiber loading from 1 to 3% in the PVA-RC biocomposite. Similar changes in elongation at break of PVA composite films have been reported previously [265,278]. Apparently, the rigid network structure of cellulose fibers limits the mobility of the chains of PVA molecules in the composite, which reduces the elongation property of the composite films.

Qazanfarzadeh and Kadivar [95] prepared biofilms based on whey protein and nanocellulose was applied as an reinforcing material. Oat husks were used as a source of cellulose material. The films under investigation were proposed as a package materials.

Nanocellulose was obtained by acid hydrolysis of oat husk cellulose (acid-base cooking) in the presence of sulfuric acid (65% H<sub>2</sub>SO<sub>4</sub>, 45 °C, 45 min, S:L of 1:8.75 (w/v)) and subsequent ultrasonic treatment (30 min). The nanocellulose particle size was 76 nm and the IC was 62.2%. The prepared nanocellulose was added in amounts of 2.5, 5 and 7.5 wt.% during the synthesis of protein isolate (WPI)-based composite film [279]. The introduction of nanocellulose into the film composition leads to an increase in its thickness from 0.119 to 0.133 mm and the formation of surface roughness, as well as a decrease in the uniformity and smoothness of the film surface. Films with a loading of nanocellulose equal to 5 wt.% showed the highest tensile strength (increase from 2.19 to 4.25 MPa), Young's modulus (increase by 47%), solubility and the lowest elongation at break and moisture content. However, the introduction of a cellulose component leads to a decrease in film transparency and water vapor permeability by 14-36%. Improvement in mechanical properties when using

polymer-based composites containing nanocellulose, with the exception of elongation, has been shown previously in the literature [280].

#### 4.5. Hydrogels and aerogels

Hydrogels are a 3D-network of hydrophilic polymers that can absorb and retain significant amounts of water and can be used in the pharmaceutical, medical, agricultural and food industries due to their excellent hydrophilicity, permeability, compatibility and low coefficient of friction [48,275,281,282]. Aerogels are a class of nanoporous materials that are of increasing interest for biomedical and pharmaceutical applications due to their porous structure and large surface area. Today, the possibility of producing hydrogels and aerogels using natural polymers, such as polysaccharides, has been shown [283,284]. Aerogels based on polysaccharides make it possible to obtain highly porous ( $\epsilon = 90-99\%$ ), lightweight (density  $0.07-0.46 \text{ g/cm}^3$ ) drug carriers with a large surface area ( $S_{\text{BET}} 70-680 \text{ m}^2/\text{g}$ ), capable ensure increased bioavailability of the drug and its carrying capacity [272,275].

Oliveira et al. demonstrated that cellulose isolated from oat husks can be applied for the production of hydrogels with poly(vinyl)alcohol [48]. To produce the target material cellulose fibers were dispersed in water (1 h,  $35 \text{ }^\circ\text{C}$ ) and added to 21% (w/v) solution of poly(vinyl)alcohol ( $90 \text{ }^\circ\text{C}$ , 1 h). The resulting mixture was stirred until being homogeneous to deliver a gel. It was supported on an acrylic plate. Hydrogels based on oat husk cellulose had low crystallinity equal to 15,4%. Swelling capacity of the obtained hydrogel and opacity were  $254.3 \pm 8.2$  and  $98.5 \pm 0.4 \%$ , respectively. The surface potential of the suspension, estimated by zeta potential analysis, of the hydrogel had a negative zeta potential of  $-12.5 \text{ mV}$ .

In [272], an aerogel based on cellulose nanocrystals and poly(vinyl)alcohol was prepared. The preparation procedure is similar to [48], but the final step was the lyophilization of the obtained hydrogel. The aerogel of oat cellulose nanocrystals had pores interconnected throughout their length. A structure was organized as a three-dimensional network which would facilitate and accelerate the diffusion of liquid into the matrix. The CI of the aerogel prepared was 79.1%. The CI value was not high for such a material. The pore size distribution, shape, pore network connectivity and CI can affect the water absorption capacity of aerogels. The prepared aerogel had a water absorption capacity equal to 402.8% due to the large pore size. In addition, it has been previously shown that nanocrystals with high relative crystallinity cause a barrier effect in polymer matrices, leading to low water diffusion between polymer chains [285]. Thus, compared with the composite material based on eucalyptus cellulose nanocrystals, which has a relative IC greater than 95%, the husk-based sample has a higher water-holding capacity. Another important characteristic is the stability of the suspension prepared from the aerogel, which is evaluated by zeta potential.

It was demonstrated in [286] that the negative charge on the surface of a suspension of materials ensures the stability of the suspension. The zeta potential reflects the surface potential of particles, which is influenced by changes to the interface with the dispersing medium, due to the dissociation of functional groups on the particle surface or adsorption of ionic species present in the aqueous dispersion medium. Thus, the stability of suspensions is determined by the strength of interaction between particles [48]. Aerogel based on nanocrystals cellulose oat hull had a zeta potential of  $-17.8 \text{ mV}$ . However, the zeta potential value of the composite based on nanocrystals of oat hull cellulose is lower than that of the composite based on cellulose extracted from rice husk. However, the use of oat husk, considered as agro-industrial waste, for the production of cellulose nanocrystals for use in aerogels has potential, for example, their use as aerogel water absorbers in food packaging.

## 5. Conclusions

The current trends in environment protection and sustainable development which are contradicted growing society's demands of materials make the investigations in the field of searching new perspective sources for any production. Oat husks seems to be a perspective resource which using could take part in the prediction of a reduction in the carbon footprint of humankind. The following advantages of oat husk may be emphasized:

- world production of oats (*Avena sativa*) reached eight million tons, it generates upto four million tons of hulls as it takes almost 50% of grain wait,
- high content of fiber in the oat husk (90 g per 100 g of the feedstock),
- cellulose amount in oat husks can be upto 45%.

In spite of significant cellulose content, it's isolation from the feedstock, purification and transformation meet some challenges justified robust and multicomponent structure, presence of lignin.

Considering the literature devoted to oat husk transformation to cellulose and the polysaccharide modification, we could conclude the following points.

1. Cellulose isolated from oat wastes can be used for the production of different materials such as hydro and aerogels, bacterial, nanocrystal and nanofibrillated cellulose, cellulose based composites and ethers. All these materials can be spent in different industries: production of pharmaceuticals, medicine (including targeted delivery of drugs, sponges etc.), agriculture (controlled delivery of pesticides or fertilizers), electronics, production of films, membranes, plastics, coatings, paints, biodegradable packages, and plenty of others.
2. Pretreatment of the oat husk biomass should be carried out in most cases. When deriving cellulose, one should take into account the following parameters of the product: the portion of impurities, the crystallinity degree and the polymerization.
3. One-step acid or alkaline treatments of oat husks at low temperatures allow one to reduce the lignin content in the composition of the cellulose product to 5-13% only. The combination of acid and alkaline treatment results in a reduction in residual non-cellulosic components in the final product. Such cellulose can be used for the synthesis of ethanol or for the production of composites based on it. Reiteration of treatment (acid-base and/or peroxide-base) combined with bleaching by bases or hydrogen peroxide produces cellulose being suitable for the synthesis of bacterial cellulose, composites, hydrogels, cellulose esters.
4. To our knowledge, only a few works are devoted to organosolv or hydrotropic cooking of oat husks. It makes investigations in this field are perspective. Additional investigations are required to increase the purity of the target cellulose.
5. A combination of chemical and physical (reactive extrusion, steam treatment) is perspective to produce pure cellulose.
6. To obtain bacterial biomass with high yield, it is necessary to use a hydrolyzate which contain only a trace amount of lignin. Pretreatment methods such as single-step alkaline or acidic are apparently not suitable for using the resulting hydrolysates for the biosynthesis of bacterial cellulose in high yields. Oat husk can be considered as a perspective source for the production of cellulose nanocrystals being used in aerogels, water absorbers, food packaging

To sum up, oat husk processing fully meets the principles of sustainable development, and it will help the environmental improvement via adopting the developed technologies.

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