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Biologically active fibers based on chitosan-coated lyocell fibers

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ABSTRACT

The possibilities of obtaining biologically active cellulose–chitosan fibers were examined. An effective two-stage method was developed. The first stage involves the formation of dialdehyde cellulose by the potassium periodate oxidation of lyocell fibers, which is able to form Schiff's base with chitosan. In the second stage, chitosan-coated lyocell fibers were prepared by subsequent treatment of oxidized lyocell fibers with a solution of chitosan in aqueous acetic acid. The impact of this two-stage protocol on the chemical and physical properties of lyocell fibers was evaluated by determining carbonyl group content, fineness and tensile strength of fibers, as well as chitosan content in the composite cellulose–chitosan fibers. Antibacterial activity of the chitosan-coated lyocell fibers against different pathogenens: *Staphylococcus aureus* and *Escherichia coli*, was confirmed *in vitro* experiments.

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1. Introduction

Over the last 10 years, the increase in the number of microbially caused diseases and hospital infections has led to intensive research into new materials, which would at the same time assure permanent biological activity together with complete safety for the customer. Therefore, there is a strong trend for searching and developing new materials which are based on the natural materials such as polysaccharides (Hon, 1996; Klemm, Heublein, Fink, & Bohn, 2005; Ravi Kumar, 2000; Strnad, Sauper, Jazbec, & Stana-Kleinschek, 2008).

Cellulose is the most abundant polysaccharide in nature and it has the potential to become a key resource in the development of sustainable biomaterials and biofuels (Klemm et al., 2005). The functionalization of cellulose fibers further broadens the potential applications of this biopolymer (Heinze & Liebert, 2001; Saito, Shibata, Isogai, Suguri, & Sumikawa, 2005; Zhang, Jiang, Dang, Elder, & Ragauskas, 2008). One of the rare examples of quite selective modification of cellulose is periodate oxidation (Calvini, Conio, Princi, Vicini, & Pedemonte, 2006; Kim & Kuga, 2000; Kim, Kuga, Wada, Okano, & Kondo, 2000; Nevell, 1957; Potthast, Kostic, Schiehser, Kosma, & Rosenau, 2007; Varma & Chavan, 1995; Varma & Kulkarni, 2002). Oxidation with periodates opens the pyranose ring and leads to the introduction of aldehydes at both C-2 and C-3 positions. On such way obtained 2,3-dialdehyde cellulose (DAC) can

be used to immobilize proteins or dyes by reaction with their amino functions, or as ion exchange materials after further oxidation of the aldehydes to the corresponding carboxylic acids (Hou, Liu, Liu, Duan, & Bai, 2008; Kim & Kuga, 2000, 2002; Zhang et al., 2008).

Chitin, a major component of the cuticles of crustaceans, insects and molluscs and the cell walls of certain fungus, is the most abundant natural amino polysaccharide with a large unexploited resources. The primary structure of chitin is similar to that of cellulose, and it may be regarded as cellulose with hydroxyl at position C-2 replaced by an acetamido group (Ravi Kumar, 2000).

Chitosan, β-(1-4) linked 2-amino-2-deoxy-β-D-glucopyranose, is prepared from chitin by deacetylation with a concentrated NaOH solution, and generally so-called chitosan or deacetylated chitin still contains acetoamino groups to some extent. Unlike cellulose, chitin and chitosan contain high percentage of nitrogen. Unique properties of these polysaccharides, such as biological activity, biocompatibility, non-toxicity and bio-resorptivity make these materials very suitable and important biomaterials which cause great industrial interest as possible substitutes for synthetic polymers. A wide variety of medical applications for chitin and chitin derivatives have been reported (Muzzarelli et al., 2007; Ravi Kumar, 2000). Recent studies in antibacterial activity of chitosan have revealed that chitosan is effective in inhibiting growth of bacteria. The antimicrobial properties of chitosan depend on its molecular weight, degree of deacetylation and the type of bacterium (Chung et al., 2004; No, Park, Lee, & Meyers, 2002).

Since chitin and chitosan fibers, due to the spontaneous crystallization of the rigid or partly rigid polymer chains, are distinctly

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brittle and crumble easily, they are not yet commercially produced on a mass scale worldwide (Mikhailov, Lebedeva, Nud'ga, & Petrova. 2001). From the other side, cellulose has been known to have good physical properties and has been widely used as construction material, paper and clothes. Due to the compatibility of two biopolymers, chitosan and cellulose, of similar structure and functions, it is possible to obtain composite fibers of defined physicomechanical properties (Liu, Nishi, Tokura, & Sakairi, 2001; Strnad et al., 2008). It has been established that chitosan reacts through its free amino groups, under specific conditions, with the aldehyde groups of oxidized cellulose giving the corresponding Schiff base. In such way, chitosan can be grafted onto the oxycellulose fibers without any synthetic cross-linking agents (Varma & Chavan, 1995). According to literature data (Chung, Lee, & Kim, 1998; Kim, Choi, & Yoon, 1998; Seong, Kim, & Ko, 1999; Strnad et al., 2008; Zhang, Chen, Ji, Huang, & Chen, 2003) much of the interest in obtaining and the antibacterial properties of cellulose-chitosan fibers has been focused on the possibilities of cotton-chitosan fiber preparation, but there is no data about using man-made cellulose fibers to produce antibacterial cellulose-chitosan fibers. In this paper, we report the potassium periodate oxidation of lyocell fibers followed by treatment with chitosan aqueous acetic acid solution to yield the chitosan-coated lyocell fibers, and their properties. Lyocell fibers were chosen for periodate oxidation because of the following: as man-made cellulose fibers they are more homogenous in structure and properties than cotton, with better sorption properties, and what is very important among man-made cellulose fibers they distinguish themselves by some unique properties (very high strength in comparison with other man-made cellulose fibers, high crystallinity, specific lustre and handle, optimum conditions for the skin (White, 2001)). Our data demonstrated that selective oxidized and chitosan-coated lyocell fibers markedly inhibit the growth of the tested pathogenens.

2. Experimental

2.1. Materials

Samples of chitosan with a Brookfield viscosity > 200 cP and deacetylation degree (DD) > 85% (denoted as H_1) and chitosan with a Brookfield viscosity > 800 cP and deacetylation degree (DD) > 75% (denoted as H_2), supplied by the Aldrich (USA), were used. Some physicochemical characteristics (moisture, nitrogen and ash content, viscosity i.e. molecular weight (M_{ν}) and deacetylation degree (DD) of chitosan samples were precisely determined and summarized in Table 1.

Lyocell fiber (fineness: 1.3 dtex, length: 38 mm; without spin finishing) was obtained from Lenzing AG, Austria. *Escherichia coli* (*E. coli*), *Staphylococcus aureus* (*S. aureus*), nutrient agar and triptone were supplied by the Institute of Virology and Immunlogy "Torlak", Belgrade. All chemicals used for the following investigations were of analytical grade.

2.2. Oxidation of lyocell fibers with potassium periodate (KIO₄)

A sample of lyocell fibers was immersed in solutions of potassium periodate in 0.1 M acetic buffer (ratio 1:50, w/v) at concentrations of 2.0 and 4.0 mg/ml, i.e. 0.2% and 0.4%, w/v. The mixture was

stirred in the absence of light, at pH 4 and room temperature, for 15, 30, 45, 60, 120, 180, 300 and 360 min. After completion of the oxidation, the lyocell fibers were washed with ice-cold water several times to remove the oxidant. These oxidized fibers were used for the chitosan coating without drying.

The effects of reaction time and periodate concentration on the rate of oxidation of cellulose fibers were studied. Consumption of one periodate molecule produces two aldehyde groups. Therefore, the rate of periodate consumption (i.e. the decrease in the periodate content of the solution, referred to the weight of fibers immersed in it, and expressed as molecules of periodate per 100 glucose units) may be identified with the rate of oxidation. Titrimetry was used to calculate the periodate consumption (Nevell. 1957).

The formation of soluble fragments, as a result of the cellulose destruction (i.e. cellulose chain scission caused by subsequent reaction, not by oxidation itself), was determined by measuring the weight loss of oxidized lyocell samples by applying the direct gravimetric method (Koblyakov, 1989).

2.3. Coating of chitosan onto the oxidized lyocell fibers

A chitosan solution was prepared in this manner: chitosan $(8.0\,\mathrm{g})$ was placed in $100\,\mathrm{ml}$ 2% (v/v) aqueous acetic acid solution for 1 h swelling and then in the dispersion of chitosan was added further $300\,\mathrm{ml}$ acetic acid solution by stirring the dispersion for 1 h at $60\,^\circ\mathrm{C}$.

The above mentioned oxidized lyocell fibers were immersed in the chitosan solution with stirring for up to 6 h, at 60 °C. After treatment, the fibers were washed with deionized water several times, and soaked in deionized water (400 ml) for 20 h at ambient temperature. The resulting fibers were dried at 60 °C for 6 h to produce the composite lyocell–chitosan fiber.

2.4. Copper number and carbonyl group content determination

Copper number of starting and periodate oxidized lyocell fibers, as a measure of the carbonyl group content, was determined according to the standard method (Tappi Test Methods, 1989–1999). A linear relationship between the carbonyl group content and copper number (Cu#) reported by Röhrling et al. (2002), as shown in Eq. (1), was used to convert the experimentally measured copper number of the oxidized lyocell fibers in carbonyl group content.

Carbonyl group content(
$$\mu mol/g$$
) = $(Cu\# - 0.07)/0.06$ (1)

2.5. Determination of fineness of oxidized lyocell fibers

Fineness in tex was determined as per standard method (SRPS F.S2.212., 1963) by dividing the mass of fibers by their know length.

2.6. Chitosan content in the composite cellulose-chitosan fibers

The chitosan content in the composite fibers was calculated from the nitrogen percentage on the basis of the calibration curve for the weight of chitosan and titration value. The nitrogen content

Table 1Some physicochemical characteristics of the chitosan samples.

Sample	Moisture (%)	Nitrogen (%)	Ash (%)	Viscosity (cP)	$M_{\rm v}$ (Da)	DD (%)
H ₁	10.79	7.46	0.76	343	252 074	87
H ₂	10.58	7.35	0.29	1 042	366 204	77

in the chitosan and cellulose-chitosan fibers was determined by Kjeldahl nitrogen analysis, according to the standard method (ISO 937, 1978), in triplicate.

2.7. Scanning electron microscopy (SEM) analysis

Scanning electron microscopy (SEM) photographs were taken on a FE-SEM JEOL JSM-6330 F instrument operating at 2 μV after sputtering with gold.

2.8. Tensile strength measurement

The tensile strength of the single fiber was measured with a Werkstoffprüfmaschinen tension tester, Germany, by following the usual procedure described elsewhere (Koblyakov, 1989). The test length of the sample fiber was 20 mm and the tensile strength of the fiber was measured as the mean of 20 individual fiber.

2.9. Assays for antibacterial activity

Gram-negative bacteria *E. coli* ATCC 25922 and Gram-positive bacteria *S. aureus* ATCC 25923 were used for antibacterial activity determination of lyocell fibers coated by chitosan. Antibacterial activity of lyocell fibers coated by chitosan was assayed as follows: each of the bacterium $(10^5-10^6 \log N/ml)$ was inoculated into 9 ml sterile potassium hydrogen phosphate buffer solution (pH 7.2) at

37 °C for 24 h. Then modified lyocell fiber samples were added in solution and incubated at 37 °C for 24 h, 48 h, 5 days and 15 days. Viable cells (log N/ml) were enumerated on TSA agar by pour plating 1 ml of serial dilutions of physiological solution followed by incubation at 37 °C for 48 h. The average values of the duplicates were converted to colony forming units per milliliter.

The percentage reduction of bacteria can be calculated by the following equation:

$$R(\%) = (A - B) \times 100/A \tag{2}$$

where *A* and *B* are bacteria amount per milliliter for the control (starting lyocell fibers) and chitosan-coated lyocell fibers test samples, respectively. In this way, by directly comparing a reference material with the treated sample, it is always possible to record the direct effect of the antimicrobial treatment, because external factors (e.g. supply of nutrients) can largely be excluded and, because of the characteristics of the sample and reference materials, it can be assumed that any potential growth pattern will be the same (Höfer, 2006).

3. Results and discussion

3.1. Obtaining of dialdehyde cellulose fibers

The first stage of obtaining of biologically active cellulosechitosan fibers involves the formation of dialdehyde cellulose by

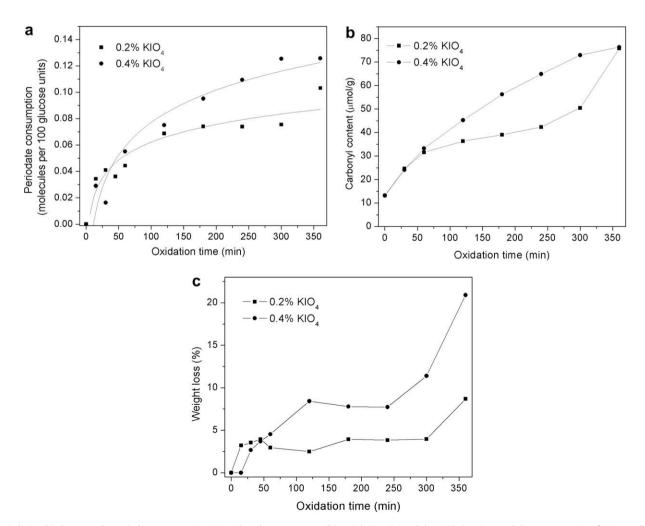


Fig. 1. Relationship between the periodate consumption (a), carbonyl group content (b), weight loss (c) and the oxidation time and the concentration of KIO₄ used for the oxidation.

the potassium periodate oxidation of lyocell fiber. The effects of periodate oxidation on lyocell fibers were initially assessed by determining the periodate consumption, carbonyl group content and weight loss.

The rate of periodate consumption by lyocell fibers from 0.2% and 0.4% potassium periodate solution is shown in Fig. 1a. The curves illustrating the course of the reaction for 0.2% and 0.4% KIO₄ show two distinct phases, i.e. the rate of periodate consumption is relatively high at first, but that it soon diminishes and becomes almost constant, Calvini and co-workers (2006) considered much larger time frames (up to 264 h) and divided the reaction course into three distinct phases: a fast initial phase with $t_{1/2}$ = 120 min, followed by a slower second reaction with $t_{1/2}$ = 20 h, and a third, rather retarded process thought to involve the oxidation of the inner core of the crystalline regions with $t_{1/2}$ = 36 d. The first reaction phase was attributed to a fast process involving the easy-to-access portion of the molecule. According to Nevell (1957) the initial fast rate of periodate consumption may be identified with complex formation. The oxidation reaction is thought to proceed via a cyclic diester of periodate with vicinal hydroxyls, which subsequently undergoes an intramolecular redox process with C-C bond cleavage according to a concerted mechanism.

As a result of periodate oxidation, lyocell fibers exhibited increase in carbonyl group content ranging from 87% to 479% with the increase of oxidation time and concentration KIO₄, Fig. 1b. During the first 60 min, no significant differences in carbonyl group content in the fibers oxidized with different concentration of KIO₄, the values are very similar. The lyocell fibers oxidized with 0.4% KIO₄ and up to 300 min had higher carbonyl group content compared to the fibers oxidized with 0.2% KIO₄. And finally lyocell fibers oxidized with 0.2% and 0.4% KIO4, during 360 min had the similar carbonyl group content with different periodate consumption. This can be explained by the fact that cellulose chain-molecules containing oxidized units are, however, sensitive and would undergo scission with the production of new end-groups and soluble fragments, as well as by periodate consumption for the overoxidation of smaller units which are part of the lost fragments. Evidence of new end groups formation can be obtained by comparison carbonyl group content against periodate consumption (Fig. 1a and b), from which we can see much higher increase in carbonyl group content than periodate consumption, especially in the case of higher periodate concentration. The formation of soluble fragments was demonstrated by measuring of the weight loss of oxidized lyocell fibers, Fig. 1c. The loss in weight of oxidized lyocell fibers is more pronounced for higher periodate charge (0.4% KIO₄), with values up to \sim 21%, while for low periodate charge these values are in the range from 3.2% to 8.6%.

The oxidation process had a significant influence on the mechanical properties of oxidized lyocell fibers. As it could be seen from Fig. 2, tensile strength of oxidized lyocell fibers decreased with increasing oxidation time. The tensile strength of the oxidized lyocell fibers did not change remarkably for the oxidation time in the range of 0–60 min. However, it significantly decreased when the oxidation time was over 60 min, probably due to breaking down the crystalline structure of cellulose, as well as remarkable increase in weight loss (see Fig. 1c). It is well known from the literature (Kim et al., 2000; Potthast et al., 2007; Varma & Chavan, 1995) that periodate attacks the crystalline regions of cellulose already at low degrees of oxidation, which affects its chemical and physical properties.

The above mentioned weight loss of the oxidized fibers followed with no or very little fiber contraction is mainly responsible for changes in the oxidized fiber surface and fineness, as it is shown in Figs. 3a, b and 4, respectively. The surface of the oxidized fiber is very rough, affected by oxidation, and some narrow lines can be

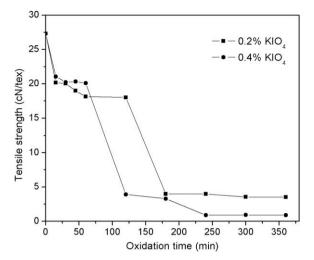


Fig. 2. The effect of oxidation time on the tensile strength of the oxidized lyocell fibers

easily seen, indicating that lyocell fibers were suffered from corrosion of periodate in the oxidation process.

After the oxidation process, the fineness of the modified fibers was increased up to 20.77%, namely from 1.30 dtex for starting (non-oxidized) fibers to 1.03 dtex for the fibers oxidized by 0.4% KIO₄ during 360 min. Higher KIO₄ concentration and increasing oxidation time increase fiber fineness. These results are in accordance with the weight loss data, and opposite to the data obtained by Princi and co-workers (2004), who observed that periodate oxidation leads to a shrinkage of cotton and linen yarns, but they worked with more than 10 times higher periodate concentration and much greater time frames (up to 120 h) than that in our study. From these data and data obtained by microscopic analysis it is clear that we obtained fibers with developed surface, that could be important for the chitosan coating.

3.2. Obtaining of chitosan-coated lyocell fibers

In the second stage, chitosan-coated lyocell fibers were prepared by subsequent treatment of oxidized lyocell fibers with a solution of chitosan in aqueous acetic acid. The free amino group of chitosan reacts with an aldehyde to give the corresponding Schiff base with high degrees of substitution.

The amount of chitosan introduced into the chemically modified lyocell fiber was determined by the Kjeldahl nitrogen analysis. Figs. 5 and 6 show the relationships between the amount of chitosan introduced into this novel fiber and the time of immersion in the chitosan solution and the reaction times of potassium periodate used for the oxidation of the cellulose fiber, respectively.

As shows Fig. 5, the amount of incorporated chitosan increased with the time of immersion in the chitosan solution (coating time) during the initial stage. When the time of immersion was over 120 min, the chitosan content became nearly constant at 0.45% for fibers oxidized by 0.2% KIO₄ during the 60 min. Therefore, chitosan coating time of 120 min were chosen for further investigations.

The aldehyde groups content of the oxycellulose reflects not only the oxidative extent of the cellulose fibers oxidized by periodate but also the extent of incorporated chitosan. The amount of incorporated chitosan increased with the oxidation time during the initial stage, whereas it decreased with longer oxidation time, Fig. 6. The chitosan content in the modified lyocell fiber was higher for fiber oxidized by 0.2% KIO₄ than fiber oxidized by 0.4% KIO₄, and for fiber immersed in the solution of chitosan with higher molecu-

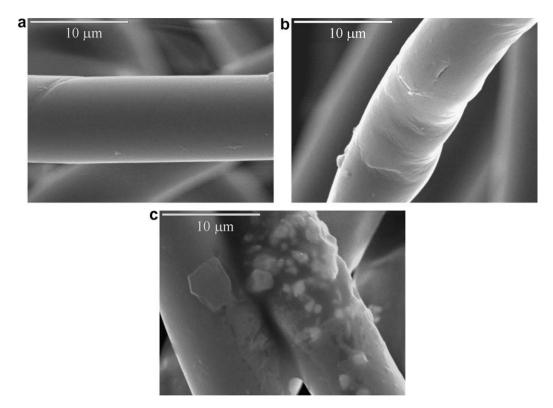


Fig. 3. SEM images of the surface of: (a) starting lyocell fiber; (b) oxidized lyocell fiber and (c) chitosan (H2) coated oxidized lyocell fiber.

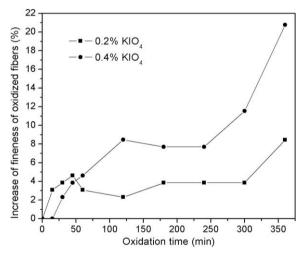


Fig. 4. The effect of oxidation conditions on the fineness of oxidized lyocell fibers.

lar weight (H_2) than chitosan with lower molecular weight (H_1) . The maximum amount of fixed chitosan was 0.51% of the weight of fibers. When the reaction time of oxidant was over 180 min, the chitosan content became nearly constant. This phenomenon can be explained by considering the differences in the reaction site of the oxidation and Schiff's base formation in the cellulose (Kim & Kuga, 2002; Liu et al., 2001). During the oxidation, the small periodate ion is able to enter the cellulose fiber interior and the glucose units inside and on the surface of the cellulose fiber can be oxidized. On the other hand, a huge chitosan molecule cannot access aldehyde groups formed in small pores of the fiber. Furthermore, a single chitosan molecule can react with many aldehyde groups.

The coating of oxidized lyocell fibers with chitosan had a significant influence on the mechanical properties of chitosan-coated

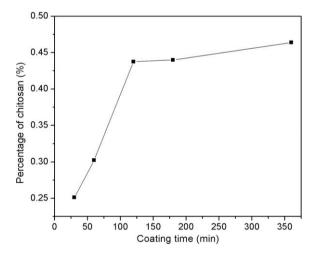


Fig. 5. Relationship between the amount of chitosan H_1 introduced into oxidized lyocell fiber (oxidation conditions: 0.2% KIO₄, 60 min) and the time of the coating by chitosan.

lyocell fibers. As it could bee seen from Table 2, tensile properties of several tested chitosan-coated lyocell fibers were lower than that of starting lyocell fibers, but the tensile strength of the chitosan-coated lyocell fibers was higher in comparison to the only oxidized lyocell fibers, therefore the chitosan acts as a coupling agent to bind fibrils to the fibers "body" and enhances the fibers mechanical properties. These can be seen from SEM images of the surface of the oxidized and the surface of the oxidized and by chitosan $\rm H_2$ coated lyocell fiber, Fig. 3. The surface of the oxidized fiber is rough, affected by oxidation, while the surface of chitosan-coated lyocell fiber is smooth, indicating that after treatment with chitosan, the surface of fibers were covered with a layer of chitosan. Also many small grains were observed on the surfaces of chitosan-coated lyocell fibers as shown in Fig. 3c.

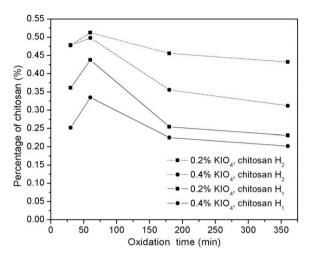


Fig. 6. Relationship between the amount of chitosan introduced into lyocell fibers and chitosan molecular weight and time of the periodate oxidation of lyocell fibers (chitosan coating conditions 120 min, 60 °C).

Table 2Tensile strength of the oxidized and chitosan-coated lyocell fibers.

Concentration of KIO ₄ (%)	Oxidation time (min)	Coated chitosan	Tensile strength (cN/tex)	CV ^a (%)
Starting lyocell fibers	0	0	27.32	11.49
0.2% KIO ₄	30	- H ₂	20.02 23.24	15.82 11.94
	180	- Н ₂	3.96 19.70	16.61 11.65
0.4% KIO ₄	60	- H ₂	20.10 19.86	9.68 16.31

^a CV-coefficient of variation.

3.3. Antibacterial activity of chitosan-coated lyocell fibers

The antibacterial properties of chitosan depend on its concentration, molecular weight, the degree of deacetylation and the type of bacteria (Chung et al., 1998; No et al., 2002). Table 3 shows the antimicrobial activity of chitosan-coated lyocell fibers. As can be seen, chitosan-coated lyocell fibers markedly inhibited growth of tested bacteria. Chitosan-coated lyocell fiber generally showed stronger bactericidal effects for Gram-positive bacteria *S. aureus*,

than for Gram-negative bacteria E. coli, which is in agreement with literature data (Hou et al., 2008; No et al., 2002). For Gram-positive bacteria S. aureus, chitosan-coated lyocell fiber with the chitosan content above 0.35% (w/w), namely samples denoted as L0-2, L60-0.4-2, L30-0.2-2 and L30-0.2-1, were the most effective, whereas for Gram-negative bacteria E. coli, sample denoted as L30-0.2-1 was the most effective. The results indicate that the lyocell fibers coated with both chitosans H₁ and H₂ show high activities against S. aureus and E. coli bacteria, whereas chitosan with higher molecular weight (H₂) is more effective than chitosan with a lower molecular weight, particularly for samples L0-2, L60-0.4-2, and L30-0.2-2. Probably, the chitosan with the higher molecular weight has the tendency to be adsorbed on the fiber surface and only slightly penetrates into the fibers. In such way, the chitosan amino groups are more accessible and able to react. Obtained results are in agreement with literature data (Zhang et al. 2003), reported that with increased molecular weight of chitosan, the reduction rate of bacteria will increase.

Samples of oxidized and chitosan-coated fibers denoted as L360-0.2-2, and L360-0.2-1, possess very low or there is no antibacterial activity (sample L360-0.2-1 against *E. coli*). Here probably, due to the long oxidation time, undesirable products of oxidation take place, which have negative influence with regard to the antibacterial activity. Also, it should be mentioned that unoxidized but chitosan-coated lyocell fibers possess antibacterial activity, but in this case chitosan is just deposited on the fiber surface without chemical binding, and stability of achieved antimicrobial activity regard to washing is very low (data not shown).

4. Conclusion

This study confirms the possibility of obtaining biologically active cellulose–chitosan fibers using oxycellulose lyocell fibers. Herein, the lyocell fibers were first oxidized by periodate to increase their aldehyde content and then chitosan was grafted onto the oxycellulose fibers without use synthetic cross-linking agents. Different periodate oxidation conditions were used to produce larger amounts of the dialdehyde cellulose or binding sites. The content of the aldehyde groups in lyocell fibers increased with increased oxidation time and with periodate concentration.

In the second stage, the free amino group of chitosan reacts with an aldehyde group of cellulose to give the corresponding Schiff base. The amount of incorporated chitosan increased with the oxidation time during the initial stage, whereas it decreased at longer oxidation time. The amount of incorporated chitosan in-

Table 3 Bacterial reduction (R, %) of chitosan coated starting and periodate oxidized lyocell fibers.

Sample code	Oxidation by KIO ₄		Chitosan type ^a	S. aureus ^b			E. coli ^c				
	Time (min)	Conc. (%)		Incubation time			Incubation time				
				24 h	48 h	5 days	15 days	24 h	48 h	5 days	15 days
LO	0	0	_	0	0	0	0	0	0	0	0
L0-2 L0-1	0	0	H ₂ H ₁	100 42.3	100 59.1	100 100	100 100	30.2 25.4	100 49.3	100 100	100 100
L60-0.4-2 L60-0.4-1	60	0.40	H ₂ H ₁	100 5.8	100 18.2	100 14.3	100 0	24.1 1.3	61.7 16.6	67.0 16.6	100 0
L30-0.2-2 L30-0.2-1	30	0.20	H ₂ H ₁	100 100	100 100	100 100	100 100	22.2 100	43.5 100	100 100	100 100
L360-0.2-2 L360-0.2-1	360	0.20	H ₂ H ₁	35.9 40.7	3.6 30.4	6.7 11.0	0 2.3	1.6 0	7.8 0	0 0	0 0

 $^{^{\}rm a}\,$ Chitosan coating conditions 120 min, 60 °C.

b Initial bacteria number 4.5 log N/ml.

^c Initial bacteria number 4.23 log N/ml.

creased with the time of immersion in the chitosan solution (coating time) during the first 120 min, and then became nearly constant. The periodate oxidation decreases the mechanical properties of fibers, while the chitosan treatment of the oxidized fibers enhances the mechanical properties of fibers, in comparison with oxidized lyocell fibers.

Chitosan-coated lyocell fiber generally shows stronger bactericidal effects for Gram-positive bacteria *S. aureus*, than for Gramnegative bacteria *E. coli*. Lyocell fiber coated with chitosans of different molecular weight show high activities against *S. aureus* and *E. coli* bacteria whereas chitosan with higher molecular weight was more effective than chitosan with a lower molecular weight.

Acknowledgments

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