Carbohydrate Polymers 79 (2010) 890–897

Contents lists available at ScienceDirect

Carbohydrate Polymers

journal homepage: www.elsevier.com/locate/carbpol

Development of anti-microbial jute fabrics via *in situ* formation of cellulose–tannic acid–metal ion complex

Asha Higazy [a,*], Mohamed Hashem [a,*], Ali ElShafei [b], Nihal Shaker [c], Marwa Abdel Hady [a]

[a] *Textile Division, National Research Centre, Dokki, Cairo, Egypt*
[b] *Division of Genetic Engineering and Biotechnology, National Research Centre, Dokki, Cairo, Egypt*
[c] *Faculty of Science, Al-Azhar University (Girls), Cairo, Egypt*

ARTICLE INFO

Article history:
Received 26 September 2009
Accepted 8 October 2009
Available online 15 October 2009

Keywords:
Anti-microbial
Cellulose
Food preservation
Jute fabric
Metal complex

ABSTRACT

Although both tannic acid and metal ions serve as anti-microbial agent, they are inactive towards several kinds of microorganisms. The current work aims at examine the technical feasibility of using tannic acid to form *in situ* complex with environmentally save metal ions onto jute cellulose. Three kind of metal salts were selected, namely, $AgNO_3$, $ZnSO_4$, $Zr(SO_4)_2 \cdot 4H_2O$. Cellulose–tannic acid–metal complexes were formed *in situ* by treatment the jute fabric with tannic acid then allow the treated samples to absorb the metal ions from its aqueous metal salt solution. The treated fabrics were monitored for anti-microbial activity and metal ion content as well as the washing durability. The effect of both tannic acid and metal salt concentration on the anti-microbial activity of the treated jute fabric as well as washing durability were investigated. Current data disclosed that jute fabrics treated with tannic acid–metal complex formed *in situ* show enhanced anti-microbial properties compared with those sample treated with tannic acid or metal ions separately and at the same concentrations. The results show also that both antibacterial and antifungal properties of the jute fabric treated with tannic–metal complexes follow the order: tannic acid—Zn > tannic acid—Zr > tannic acid—Ag

It was found also that the washing durability of jute fabric treated with tannic acid–metal complex was very high and depends on the type of metal ion used in complexation and follows the order:

tannic acid—Zn > tannic acid—Ag > tannic acid—Zr.

1. Introduction

Anti-microbials gain interest from both academic research and industry due to their potential to provide quality and safety benefits to many materials. Anti-microbial packaging is the packaging system that is able to kill or inhibit spoilage and pathogenic microorganisms that are contaminating foods (Church & Parsons, 2007; Devlieghere, Vermeiren, Jacobs, & Debevere, 2000; El-Refaie, Worley, & Broughton, 2007; Ghosh, Srivatsa, Nirmala, & Sharma, 1977; Han, 2003; Weng & Hotchkiss, 1993).

Microbial contamination reduces the shelf life of foods and increases the risk of food-borne illness. The demand for minimally processed, easily prepared, and ready to eat "fresh" food products, poses major challenges for food safety and quality. Application of anti-microbial treatment in food packaging is gaining interest from researchers due to its potential to provide quality, safety benefits and to extend the shelf life of the food. Anti-microbial food packaging promotes safety by reducing the rate of growth of specific microorganisms by direct contact of the package with the surface of foods (Coma, Sebti, Pardon, Deschamps, & Pichavant, 2001; Ming, Weber, Ayres, & Sandine, 1997; Weng & Hotchkiss, 1993).

The ideal anti-microbial polymer should possess the following characteristics: (i) easily and inexpensively synthesized, (ii) stable in long-term usage and storage at the temperature of its intended application, (iii) not soluble in water for a water disinfection application, (iv) does not decompose to and/or emit toxic products, (v) should not be toxic or irritating to those who are handling it, (vi) can be regenerated upon loss of activity, and (vii) biocidal to a broad spectrum of pathogenic microorganisms in brief times of contact (Church & Parsons, 2007; Jay, 1996; Scannell et al., 2000).

A tremendous effort has been made over the last decade to develop and test films with anti-microbial properties to improve food safety and shelf life. Some polymers are inherently anti-microbial, and have been used in films and coatings. Cationic polymers such as chitosan promote cell adhesion. Because charged amines interact with negative charges on the cell

* Corresponding authors. Fax: +20 233370931.
E-mail addresses: gelfeky@hotmail.com (A. Higazy), mmhashem@ncsu.edu (M. Hashem).

0144-8617/$ - see front matter © 2009 Elsevier Ltd. All rights reserved.
doi:10.1016/j.carbpol.2009.10.019

membrane, they can cause leakage of intracellular constituents. Chitosan has been used as a coating and appears to protect fresh vegetables and fruits from fungal degradation (Chung, Papadakis, & Yam, 2003; Han & Floros, 1998; Joerger, 2003; Liu et al., 2007; Plackett, Holm, & Johansen, 2006).

For food preservation, chitosan films are very effective. Chitosan has widely been used in anti-microbial films, to provide edible protective coating, dipping and spraying for the food products due to its anti-microbial properties. Chitosan films have been used as a packaging material for the quality preservation of a variety of food. Chitosan has great potential for a wide range of applications due to its biodegradability, biocompatibility, anti-microbial activity, non-toxicity and versatile chemical and physical properties (Chen, Yeh, & Chiang, 1996; Ouattara, Simard, Piette, et al., 2000).

It is possible that research and development of "intelligent" or smart anti-microbial food packages will follow. These will be materials that sense the presence of microorganisms in the food.

Tannins are present in a variety of fruits and vegetables (Deshpande, Sathe, & Salunkhe, 1984). Wine and tea also contain tannins. Vegetable tannins are water-soluble phenolic compounds having molecular weight between 500 and 3000 Da. Vegetable tannins are classified into hydrolysable tannins and condensed tannins. Hydrolysable tannins contain either gallotannin or ellagitannins. Condensed tannins are the polymerized products of flavan-3-ols and flavan-3,4-diols or mixture of the two polymers, referred to as "flavans" (Salunkhe, Chavan, & Kadan, 1989; Sanderson et al., 2001).

Tannins acts like mild acid on the basis of many phenolic–OH groups. Tannic acid is the simplest form of hydrolysable tannin. High quality tannin contains 65–76% tannic acid. One of the most important properties of tannins and tannic acid is their strong ability to form chelating complexes with metal ion. Metal complexation properties of tannins and tannic acid are used nowadays in certain textile dyeing and tanning. Although tannins and tannic acid may serve as natural anti-microbial agents, they are inactive towards a broad spectrum of fungi and bacteria (Salunkhe et al., 1989; Sanderson et al., 2001).

However, to the author's knowledge, no work in the literature describes the anti-microbial activity of jute fabric treated with tannic acid–metal complex. This indeed stimulates the present work. The aim of our work is to study the technical feasibility to impart an anti-microbial activity to jute fabric using *in situ* formation of tannic acid–metal ion. Tannic acid–metal ion complexes were formed *in situ* by first treatment jute fabric with tannic acid then allow the treated samples to absorb the metal ion from its aqueous metal salt solution. The treated fabrics were monitored for anti-microbial activity and metal ion content. For this purpose, three kind of metal salts were investigated, namely, $AgNO_3$, $ZnSO_4$, $Zr(SO_4)_2{\cdot}4H_2O$. The effect of both tannic acid and metal salt concentration on the anti-microbial activity of the treated jute fabrics were also investigated; the washing durability was also examined.

2. Experimental

2.1. Materials

Grey jute fabric was kindly supplied from Jute Company, Cairo, Egypt. The fabric was scoured using an aqueous solution containing NaOH, 40 g/l, Egyptol®, 5 g/l, at 95 °C for 30 min. The fabric was then washed several times with boiling water then washed with cold water and finally dried at ambient conditions.

Silver nitrate, zinc sulphate, zirconium sulphate tetrahydrate and tannic acid were of laboratory grade chemicals. Egyptol® (non-ionic wetting agent based on ethylene oxide condensate) was of commercial grade chemicals.

2.1.1. Scouring of jute fabric

Grey jute fabric was scoured using an aqueous solution containing NaOH, 40 g/l; Egyptol, 5 g/l; at 95 °C for 30 min. The fabric was then washed several times with boiling water then washed with cold water and finally dried at ambient conditions.

2.1.2. Treatment of jute fabric using in situ formation of tannic acid–metal ion complex

A known weight of tannic acid was dissolved in distilled water. Scoured jute fabrics were padded in two dip and nip in tannic acid solution then squeezed to a wet pick-up of 100%. The jute fabrics were then dried at 85 °C for 5 min. The sample was then padded in an aqueous solution containing known concentration of different metal salts, namely silver nitrate [$AgNO_3$], zinc sulphate [$ZnSO_4$] and zirconium sulphate [$Zr(SO_4)_2{\cdot}4H_2O$] and then squeezed to a wet pick-up 100%. The samples were then dried at 85 °C for 5 min. At this end, the samples were washed several times with distilled water at room temperature and finally dried at ambient conditions.

2.2. Testing and analysis

2.2.1. Anti-microbial activity test

Control and treated jute samples were tested for their anti-microbial activities against *Staphylococcus aureus* as Gram positive bacteria and *Candida albicans* as a fungus. Microorganisms were supplied from the Department of Microbial Chemistry, Division of Genetic Engineering and Biotechnology, National Research Center, Cairo, Egypt. The cultural medium used was prepared by mixing the following constituents: glucose, 10 g/l; yeast extract, 3 g/l; meat extract, 1.5 g/l; NaCl, 0.5 g/l; and agar 20 g/l. The pH of the cultural medium was adjusted at 7 then sterilized at 120 °C for 30 min under pressure (Koneman, Allen, Jonda, & Win, 1994, chap. 19).

Anti-microbial activity was estimated according to AATCC standard test method (AATCC Standard Test method No. 90-1977). The previously mentioned medium was poured in sterile Petri dishes (20 ml for each plate) and left to cool. These plates were inoculated with the test organism and left for 2 h. Discs of jute fabric samples (10 mm in diameter) were introduced to the plates with a sterile forceps and gently pressed to insure good contact with the solid medium. The plates were then kept in the refrigerators at 5 °C for 1 h to permit good diffusion before transferring them to an incubator at 37 °C for 24 h. The inhibition zones (mm) were then measured.

2.2.2. Metal ion determination

Metal ion content of jute fabric before and after treatment with chitosan–metal complex as well as those treated with metal salts were evaluated using "Flame Atomic Absorption Spectrometer", Varian Spectra A220. The experimental technique was adopted as follows: 0.5 g of dried jute fabric was dissolved in 50 ml 72% H_2SO_4 at 3 °C. The solution was completed to 250 ml using distilled water. 10 ml from this solution was diluted to 500 ml using distilled H_2O. Aliquot from the latter was subjected to "Flame Atomic Adsorption" determination. The metal ion content [C (ppm)] of jute fabric was determined from the following equation:

$$C(\mathrm{ppm}) = 2500\ A$$

where, A = is the value obtained from flame atomic adsorption spectrophotometer.

3. Results and discussion

Tannic acid was found to be inhibitory to the growth of broad spectrum of bacteria and fungi. However, tannic acid was found to be inactive towards the growth of several other bacteria such as *Bifidobacterium infantis* and *Lactobacillus acidophilus* (Salunkhe et al., 1989; Sanderson et al., 2001). Tannic acid has high binding efficiency to heavy metal (Salunkhe et al., 1989). The work presented in this part deal with the anti-microbial finishing of jute fabric by an easy, cost effective method with laundering durability. For this, we attempt to adsorb three kinds of metallic salts onto jute fabric after treatment with tannic acid then examine their anti-microbial activity against *S. aureus* and *C. albicans*. The washing durability of the treated samples was also examined. Tannic acid–metal complex was formed *in situ* inside the jute fabric by first treated fabric with an aqueous tannic acid solution of known concentration at room temperature, and then squeezed to wet pick-up of 100%, dried at 85 °C for 5 min followed by treatment with aqueous metal salt solution of known concentration then squeezed to wet pick-up of 100% dried at 85 °C for 5 min. At this end, the fabric washed with distilled water and dried at ambient conditions.

Three metal cations were allowed separately to form complex with jute fabric pretreated with tannic acid. These are, silver as silver nitrate, zinc as zinc sulphate and zirconium as zirconium sulphate. The effect of metal salt type and its concentration as well as tannic acid concentration on the anti-microbial activity of jute fabric were studied. Results obtained along with the appropriate discussion are given below:

3.1. Effect of silver nitrate and tannic acid concentrations on the anti-microbial properties of thereof treated jute fabrics

Table 1 shows the effect of silver nitrate and tannic concentrations on the anti-microbial properties of the treated jute fabric. The anti-microbial properties were evaluated by measuring the inhibition zone formed around the test sample as described in the experimental section. *S. aureus* and *C. albicans* were used as examples of bacteria and fungi, respectively. Anti-microbial properties of jute samples treated with different concentrations of tannic acid only are set-out in the same table under zero $AgNO_3$ concentration. Also the anti-microbial properties of jute sample treated with $AgNO_3$ only are set-out in the same table under zero tannic acid concentration. Results of Table 1 reveal that:

(i) All jute samples treated with tannic acid only (i.e., at zero $AgNO_3$) show anti-microbial properties, which vary according to the concentration of tannic acid used. It is further noted that increasing the tannic acid concentration from 0.25% to 1.5% is accompanied by an increases in the inhibition zone from 1.3 and 2.5 mm to 4 mm and for *S. aureus* and *C. albicans*, respectively. Further increase in tannic acid concentration exerts practically no effect on the anti-microbial properties of jute fabric. The same holds true for jute fabric treated with different concentrations from silver nitrate only (i.e., at zero concentration of tannic acid). The inhibition zone increases from 1 to 3.5 mm for *S. aureus* to *C. albicans*, respectively, to 4 mm for both *S. aureus* and *C. albicans* by increasing $AgNO_3$ from 0.05% to 0.25%.

(ii) At constant concentration from tannic acid, increasing $AgNO_3$ concentration enhances the anti-microbial properties of jute fabric. This is evidenced by the substantial increase in the inhibition zone. It is also observed that, at constant $AgNO_3$ concentration, the higher tannic acid concentration the higher improvement in anti-microbial properties of jute as evidenced by increase in inhibition zone.

Results of Table 1 feature that, higher antifungal and antibacterial properties were attained when the jute fabric was first treated with 1.5% tannic acid followed by treatment with 0.15% $AgNO_3$. Current data disclosed that, mean while both tannic acid and $AgNO_3$ serve as anti-microbial agent for jute fabric, *in situ* complex formation of tannic acid–Ag complex inside the jute fabric enhances the anti-microbial properties of the latter, even at low concentration from tannic acid and $AgNO_3$.

3.2. Effect of zinc sulphate and tannic acid concentrations on the anti-microbial activity of thereof treated jute fabric

Table 2 shows the dependence of the anti-microbial properties of jute fabric on the concentrations of both tannic acid and zinc sulphate. The results signify that, the anti-microbial activity of jute fabric treated with tannic acid–Zn complex formed *in situ* on the surface and inside the jute fabric is directly related to the concentration of both tannic acid and zinc sulphate. The trend obtained are rather similar to that obtained above with tannic acid–Ag complex and could be explained on similar basis.

A comparison between the results of Table 1 and those of Table 2 would conclude main point. At the same concentration of tannic acid, $AgNO_3$ and $ZnSO_4$, the anti-microbial properties of the jute fabric treated with tannic acid–Zn complex are higher than that obtained with tannic acid–Ag complex.

3.3. Effect of zirconium sulphate and tannic acid concentrations on the anti-microbial properties of thereof treated jute fabric

Table 3 shows the dependence of the anti-microbial properties of jute fabric on the concentrations of both tannic acid and zirconium sulphate. The results signify that the anti-micro-

Table 1
Effect of silver nitrate and tannic acid concentrations on the anti-microbial activity of treated jute fabric.

Type of microorganism	$AgNO_3$ conc. (%)	Inhibition zone (mm)					
		Tannic acid concentration (%)					
		0.00	0.25	0.50	1.00	1.50	2.00
S. aureus	0.00	0.00	1.30	2.50	3.50	4.00	4.00
	0.05	1.00	1.80	3.00	3.88	4.50	4.90
	0.10	2.00	2.50	3.50	4.00	4.80	5.20
	0.15	2.50	3.80	4.50	5.00	6.50	6.50
	0.25	4.00	4.50	5.00	5.00	6.50	6.00
C. albicans	0.00	0.00	2.50	3.00	3.50	4.00	4.00
	0.05	3.50	3.80	4.00	4.50	4.50	5.00
	0.10	3.80	4.50	4.50	5.00	5.50	5.50
	0.15	4.00	5.00	5.00	5.50	6.00	6.00
	0.25	4.00	5.00	5.50	5.50	6.00	6.00

Conditions used: drying temperature at 85 °C and drying at time 5 min.

Table 2
Effect of zinc sulphate and tannic acid concentrations on the anti-microbial activity of treated jute fabric.

Type of microorganism	$ZnSO_4$ conc. (%)	Inhibition zone (mm)					
		Tannic acid concentration (%)					
		0.00	0.25	0.50	1.00	1.50	2.00
S. aureus	0.00	0.00	1.30	2.50	3.50	4.00	4.00
	0.05	3.50	4.50	4.80	5.50	5.60	5.60
	0.10	4.00	5.70	5.80	6.50	7.00	7.00
	0.15	6.00	6.80	7.20	7.80	8.50	8.50
	0.25	7.00	7.50	7.90	8.00	8.50	8.50
C. albicans	0.00	0.00	2.50	3.00	3.50	4.00	2.00
	0.05	2.00	3.00	4.00	4.50	4.50	4.50
	0.10	3.00	3.50	4.50	5.00	5.50	5.50
	0.15	4.00	5.00	5.50	6.00	6.50	6.50
	0.25	5.00	5.00	6.00	6.50	6.50	6.50

Conditions used: drying temperature at 85 °C and drying at time 5 min.

Table 3
Effect of zirconium sulphate and tannic acid concentrations on the anti-microbial activity of treated jute fabric.

Type of microorganism	$Zr(SO_4)_2 \cdot 4H_2O$ conc. (%)	Inhibition zone (mm)					
		Tannic acid concentration (%)					
		0.00	0.25	0.50	1.00	1.50	2.00
S. aureus	0.00	0.00	1.30	2.00	2.50	3.50	4.00
	0.05	1.50	2.00	2.50	2.80	3.80	4.50
	0.10	3.50	4.00	4.30	4.50	5.00	5.00
	0.15	4.00	4.50	4.50	5.00	7.50	7.50
	0.25	4.00	5.20	5.50	5.50	7.00	7.00
C. albicans	0.00	0.00	2.50	3.00	3.00	3.50	4.00
	0.05	2.50	3.00	3.20	3.50	4.00	4.50
	0.10	3.00	4.00	4.50	4.80	5.50	5.50
	0.15	4.00	4.80	5.00	5.00	6.50	6.50
	0.25	4.00	5.20	5.50	5.50	7.00	7.00

Conditions used: drying temperature at 85 °C and drying time at 5 min.

Table 4
Percent loss of anti-microbial properties and metal ion content after repeated wash.

Metal ion	After treatment (before washing)			After 5 washing cycles			After 10 washing cycles			After 15 washing cycles		
	I.Z (mm)		$[M^+]$ (ppm)	I.Z (mm)		$[M^+]$ (ppm)	I.Z (ppm)		$[M^+]$ (ppm)	I.Z (mm)		$[M^+]$ (ppm)
	S. aureus	*C. albicans*		*S. aureus*	*C. albicans*		*S. aureus*	*C. albicans*		*S. aureus*	*C. albicans*	
Ag^+	0.0	0.0	0.0	4.6	3.3	3.9	7.7	8.3	5.9	10.7	8.3	5.9
Zn^{2+}	0.0	0.0	0.0	3.75	4.6	3.5	5.9	4.6	4.7	5.9	4.6	5.3
Zr^{2+}	0.0	0.0	0.0	6.6	7.6	5	10.6	10.2	7.5	13.3	10.8	10

I.Z = Inhibition zone (mm).
Conditions used: [tannic acid], 2%; [metal salt], 0.15%; drying temperature, 85 °C; drying time, 5 min.

bial activity of jute fabric treated with tannic acid–Zr complex formed *in situ* inside and on the surface of the jute fabrics is directly related to the concentrations of both tannic acid and zirconium sulphate. The trends obtained are rather similar to those obtained above with both tannic acid–Zn and tannic acid–Ag complexes and could be explained on similar basis.

A comparison between the results of Tables 1–3 would conclude that, at the same tannic acid and metal concentration, the anti-microbial properties of jute fabric treated with tannic acid–metal complex follow the order:

$$\text{tannic acid—Zn} > \text{tannic acid—Zr} > \text{tannic acid—Ag}$$

3.4. Washing durability of jute fabric treated with tannic acid–metal complex

Although tannic acid–metal complexes are water insoluble complexes, it is very important to examine the durability of these finishing treatments to several washing cycles. Hence, jute samples treated with silver, zinc or zirconium ions complexed with tannic acid were subjected to several washing cycles using distilled water and wetting agent (non-ionic wetting agent). The samples were then dried and subjected to both anti-microbial activity tests and metal ion content by processes similar to that mentioned previously. The results obtained just after finishing treatment before washing and after 5, 10 and 15 washing cycles are set-out in Table 4 and Figs. 1–3. It is seen from Table 4 and Figs. 1–3 that:

(i) The antibacterial activities of jute fabric samples were maintained even after 15 washing cycle although there is marginal decrement in the inhibition zone. It is further observed that the washing durability of the antibacterial properties of the jute fabric treated with tannic acid–metal complex follow the order:

Tannic acid–Zn > tannic acid–Ag > tannic acid–Zr. The same order is also observed for the antifungal durability of jute fabric against several washing.

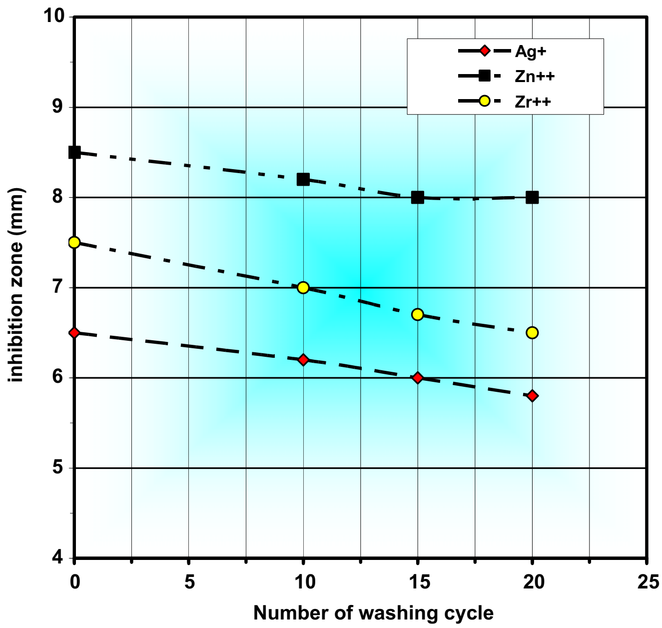


Fig. 1. Effect of repeated washing on anti-microbial properties of jute fabric treated with tannic acid–metal complex. Conditions used: [tannic acid], 2%; [metal salt], 0.15%; drying temperature, 85 °C; drying time, 5 min.

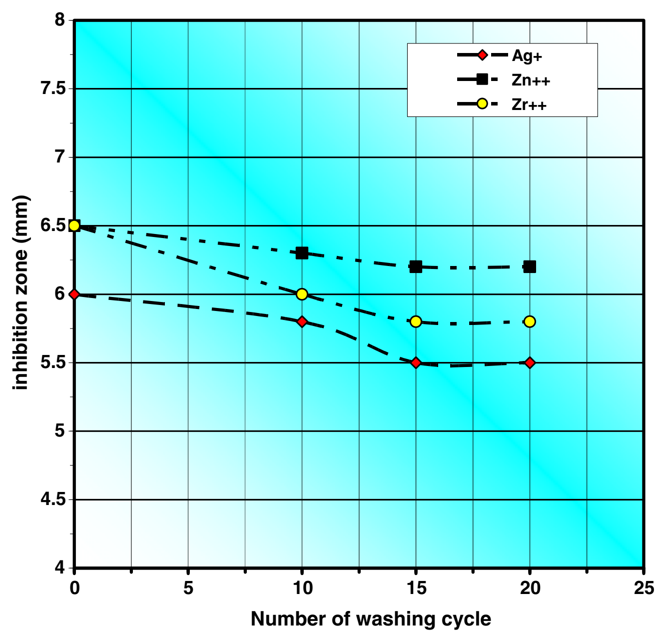


Fig. 2. Effect of repeated washing on antifungal properties of jute fabric treated with tannic acid–metal complex. Conditions used: [tannic acid], 2%; [metal salt], 0.15%; drying temperature, 85 °C; drying time, 5 min.

(ii) The metal ion content of the treated jute fabric samples were maintained even after 15 washing cycles with different degree of stability. Jute sample treated with tannic acid–Zr complex shows the lowest stability, where the % loss of metal ion in its concentration after 15 washing cycles was amounted 10%. The highest washing durability was observed with those samples treated with tannic acid–Zn complex where the % loss does not exceed 5.3% after 15 washing cycles.

The results of Table 4 and Figs. 1–3 show that, the washing durability of jute fabric treated with tannic acid–metal complexes

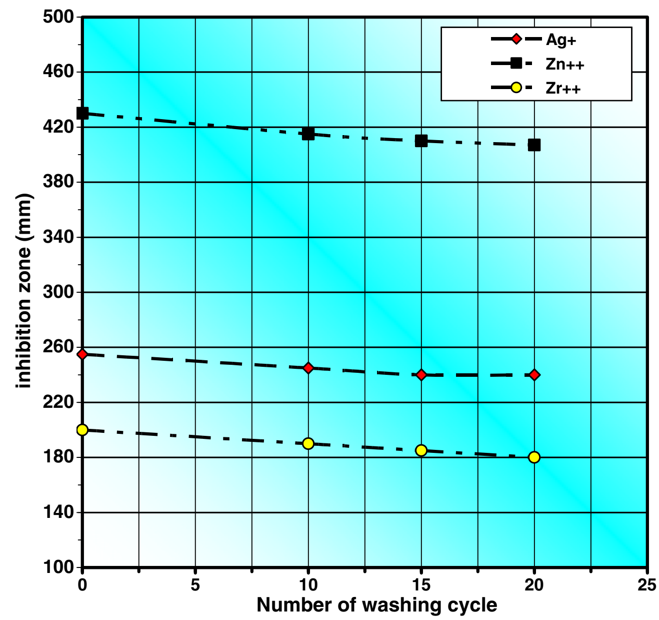


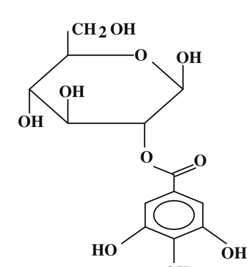
Fig. 3. Effect of repeated washing on metal ion content of jute fabric treated with tannic acid–metal complex. Conditions used: [tannic acid], 2%; [metal salt], 0.15%; drying temperature, 85 °C; drying time, 5 min.

depended on the type of metal ion used in complexation with tannic acid and follow the order:

tannic acid—Zn > tannic acid—Ag > tannic acid—Zr.

3.5. Proposed mechanism for formation of tannic acid–metal cation–cellulose complex

Certain structure and configurations of individual molecules enable them to coordinate with metal ions not only behaves ligand like one but also like two, three and four ligands. Such ligand known as polydentate ligand. Tannic acids (Structure 1) with its three phenolic groups consider a polydentate ligand.

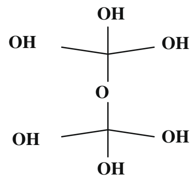


Structure (I) Tannic acid

"Composed of one glucose molecule and one galloyl group"

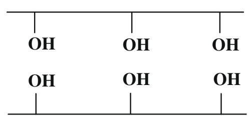
Presumably, metal ions can coordinate with the adjacent hydroxyl on the galloyl groups and/or hydroxyl groups in glucose residue. This is illustrated by scheme 1. Both intra- and/or inter-chelating bond are formed between tannic acid and metal ions. Inter-chelating bond formed between the two adjacent hydroxyl groups in galloyl group and the metal ions form the more stable five member rings.

For simplicity, the structure of tannic acid will be represented as follows:



Schematic representation of tannic acid

And that for jute cellulose will be represented as follows:



Schematic representation of cellulose

Scheme 2 depicts the formation of tannic acid–metal ion cellulose complex. Jute fabric (and generally cellulosic fabric) adsorbs tannic acid from its aqueous solution. In this case, the interaction between tannic acid and cellulose is mainly hydrogen bonding and minor Van-der-Wall interaction. When the treated jute fabric with tannic acid is further treated with a solution of metal salt, in subsequent step, a fast and stable complex is formed between both tannic acid and cellulose with metal ions impeded in cellulose chain through an intra/inter coordinate bond as explained previously.

The good washing fastness properties of jute fabric treated with tannic acid–metal complex is attributed to two reasons:

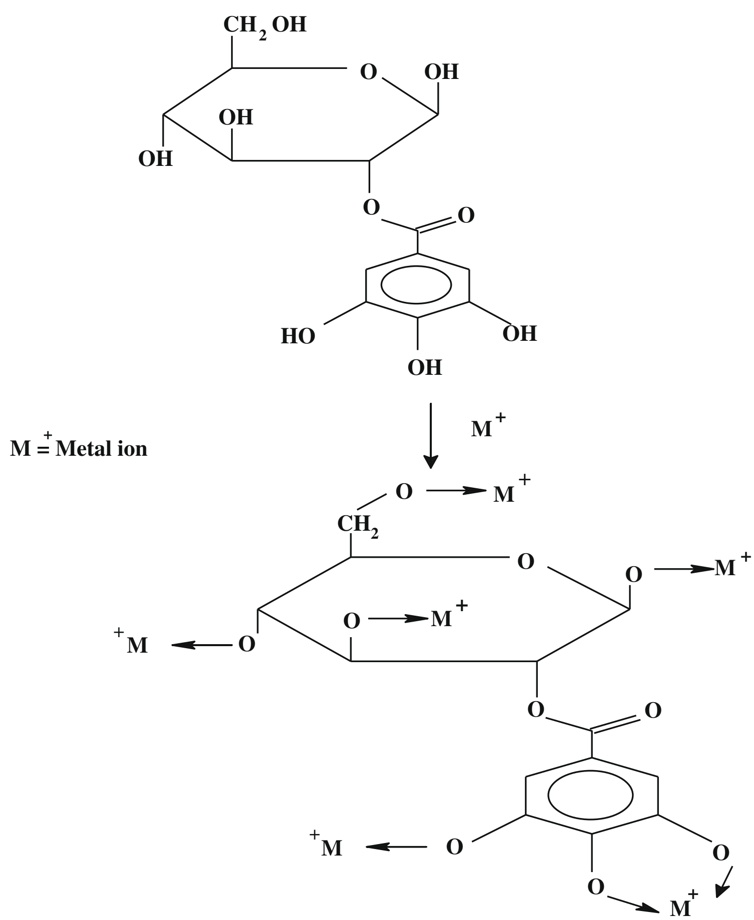
(i) The strong coordinate bond formed between metal ions, tannic acid and hydroxyl group of cellulose molecules.
(ii) The formation of large insoluble complex molecules impeded in cellulose macromolecules. These two factors contribute to more or less extent in enhancement the washing fastness of jute fabric treated with tannic acid–metal complex. Results obtained previously indicate also that the washing fastness of jute fabric treated with tannic acid–metal complex depends on the type of metal ion and its concentrations as well as the concentration of tannic acid (polydentate ligand).

4. Conclusions

Current data disclosed that jute fabrics treated with tannic acid–metal complex formed *in situ* show enhanced anti-microbial properties compared with those sample treated with tannic acid or metal ions in uncomplexed form and at the same concentration of both. The results show also that both antibacterial and antifungal properties of the jute fabric treated with tannic–metal complexes follow the order:

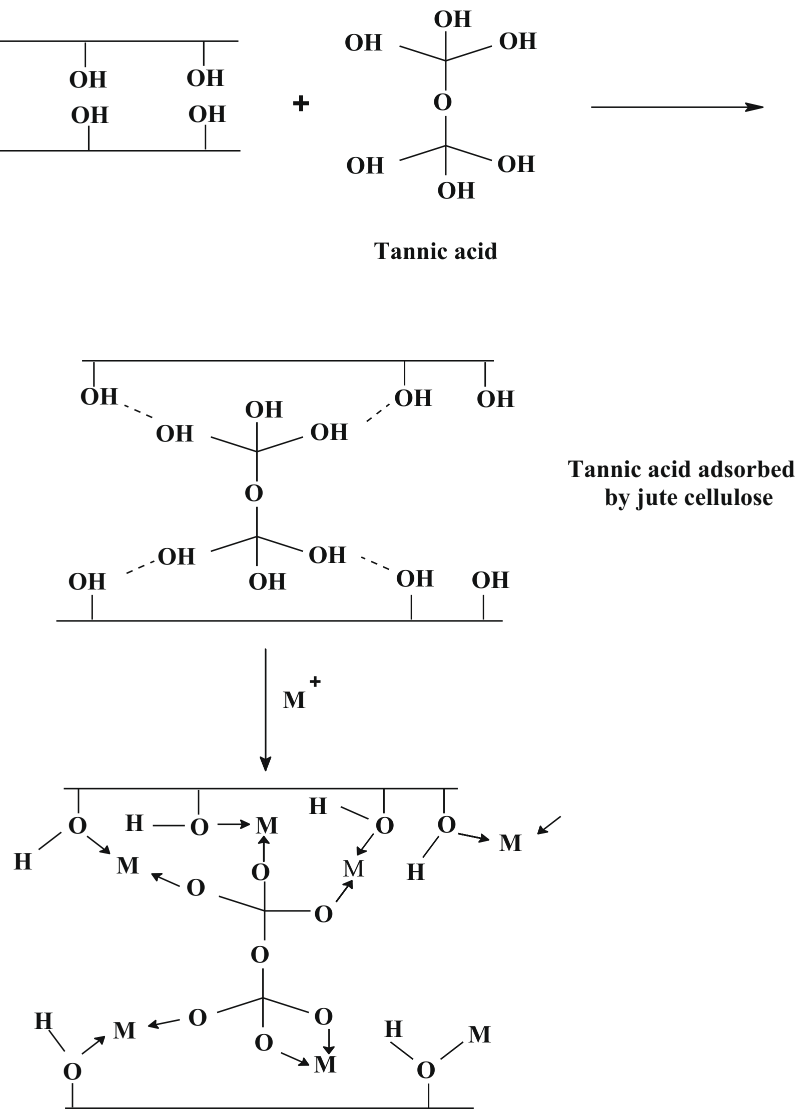
tannic acid—Zn > tannic acid—Zr > tannic acid—Ag

It was found also that the washing durability of jute fabric treated with tannic acid–metal complex was very high compared with that



Tannic acid metal ion complexes

Scheme 1. Formation of tannic acid–metal ion complex.



Scheme 2. Formation of tannic acid–metal ion cellulose complex.

obtained with chitosan–metal complexes at the same type metal ion. Washing durability of jute fabric treated with tannic acid–metal complexes depends on the type of metal ion used in complexation and follow the order:

tannic acid—Zn > tannic acid—Ag > tannic acid—Zr

Acknowledgment

The authors acknowledge the Late – Prof. Dr. Osama Mohamed Abdel Fatah; Division of Genetic Engineering and Biotechnology, National Research Centre, Cairo, Egypt, for his technical support, sound advices and sincere help throughout this work.

References

AATCC Standard Test method No. 90-1977.
Chen, M., Yeh, G., & Chiang, B. (1996). Antimicrobial and physicochemical properties of methylcellulose and chitosan films containing a preservative. *Journal of Food Processing and Preservation, 20*, 379–390.
Chung, D., Papadakis, S. E., & Yam, K. L. (2003). Evaluation of a polymer coating containing triclosan as the antimicrobial layer for packaging materials. *International Journal of Food Science and Technology, 38*, 165–169.
Church, I., & Parsons, A. (2007). Antimicrobial films for food applications: A quantitative analysis of their effectiveness. *Packaging Technology and Science, 20*, 4, 231–273.
Coma, V., Sebti, I., Pardon, P., Deschamps, A., & Pichavant, F. H. (2001). Antimicrobial edible packaging based on cellulosic ethers, fatty acids, and nisin incorporation to inhibit *Listeria innocua* and *Staphylococcus aureus*. *Journal of Food Protection, 64*, 470–475.
Deshpande, S., Sathe, S., & Salunkhe, D. (1984). Chemistry and safety of plant polyphenols. *Advances in Experimental Medicine and Biology, 177*, 457.
Devlieghere, F., Vermeiren, L., Jacobs, M., & Debevere, J. (2000). The effectiveness of hexamethylenetetramine incorporated plastic for the active packaging of foods. *Packaging Technology and Science, 13*, 117–121.
El-Refaie, K., Worley, S., & Broughton, R. (2007). The chemistry and applications of antimicrobial polymers: A state-of-the-art review. *Biomacromolecules, 8*(5), 1359.
Ghosh, K., Srivatsa, A., Nirmala, N., & Sharma, T. (1977). Development and application of fungistatic wrappers in food preservation. Part II. Wrappers made by coating process. *Journal of Food Science and Technology, 14*, 261–264.
Han, J. H. (2003). Antimicrobial food packaging. In R. Ahvenainen (Ed.), *Novel food packaging techniques* (pp. 50–70). Boca Raton, FL: CRC Press LLC.
Han, J. H., & Floros, J. D. (1998). Simulating diffusion model and determining diffusivity of potassium sorbate through plastics to develop antimicrobial packaging films. *Journal of Food Processing and Preservation, 22*, 107–122.
Jay, J. M. (1996). *Modern food microbiology* (5th ed., pp. 291–293). New York, NY: Chapman & Hall.
Joerger, R. (2003). Inhibition of microbial growth in ready-to-eat food stored at ambient temperature by modified atmosphere packaging. *Packaging Technology and Science, 16*(6), 239–247.
Koneman, E., Allen, S., Jonda, S., & Win, Jr., (1994). *Introduction to diagnostic microbiology* (5th ed., p. 785). Philadelphia: Lippincott.

Liu, L., Finkenstadt, V., Liu, C., Jin, T., Fishman, M., & Hicks, K. (2007). Preparation of poly(lactic acid) and pectin composite films intended for application in antimicrobial packaging. *Journal of Applied Polymer Science, 106*, 801–810.

Ming, X., Weber, G., Ayres, J., & Sandine, W. (1997). Bacteriocins applied to food packaging materials to inhibit *Listeria* monocytogenes on meats. *Journal of Food Science, 62*, 413–415.

Ouattara, B., Simard, R. E., Piette, G., et al. (2000). Diffusion of acetic and propionic acids from chitosan based antimicrobial packaging films. *Journal of Food Science, 65*, 768–773.

Plackett, D. V., Holm, V. K., & Johansen, P. (2006). Characterization of l-polylactide and l-polylactide-polycaprolactone co-polymer film for use in cheese-packaging applications. *Packaging Technology and Science, 19*, 1–24.

Salunkhe, D., Chavan, J., & Kadan, S. (1989). *Nutritional consequence of dietary tannins in "Dietary Tannins": Consequents and remedies*. Boca Raton, FL: CRC Press. p. 113.

Sanderson, G., Ranadive, A., Eisenburg, L., Farrel, F., Simons, R., Manley, C., et al. (2001). Contribution of polyphynolic compounds to the taste of tea, sulfur and nitrogen compound in food flavors. In G. Charalambous, I. Katz (Eds.), *Acs symposium series* (Vol. 26, p. 14). Washington, DC: American Chemical Society.

Scannell, A., Hill, C., Ross, R., Marx, S., Hartmeier, W., & Arendt, E. (2000). Development of bioactive food packaging materials using immobilized bacteriocins Lacticin and Nisaplin. *International Journal of Food Microbiology, 60*, 241–249.

Weng, Y.-M., & Hotchkiss, J. H. (1993). Anhydrides as antimycotic agents added to polyethylene films for food packaging. *Packaging Technology and Science, 6*, 123–128.