

HYGIENIC PAPERS MODIFIED BY FUNCTIONAL BIOPOLYMER-BIOCIDE COMPOSITIONS IN A PILOT RESEARCH INSTALLATION

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Abstract

The aim of this work was to demonstrate the process of manufacturing and applying functionalising additives using a pilot research installation. As part of the optimisation evaluation, hygienic papers functionalised with biopolymer-biocide compositions based on chitosan, starch and Gemini surfactants were prepared. The microbiological properties and susceptibility to biodegradation of prototype hygienic papers (prepared in EPICOM) were assessed. In particular, the minimum amount of biopolymer and bioactive agent were determined to ensure the finished product exerted antibacterial activity against *Staphylococcus aureus*, *Bacillus subtilis* and *Escherichia coli* and was more than 90% biodegradable in a compost environment. Under optimal conditions, the hygienic paper exerted excellent antibacterial activity against *S. aureus* and *E. coli* as well as good activity against *B. subtilis*. Moreover, the hygienic paper showed more than 90% biodegradability in compost conditions within 8 weeks.

Keywords: biodegradable, bioactive hygienic papers, Gemini surfactants, chitosan-starch functional compositions

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1. Introduction

Washing hands with soap and drying them with a disposable paper towel is the method recommended by the World Health Organization (WHO) to remove germs. The present study aimed to develop an innovative hygienic paper based on an in-depth analysis of the needs and expectations of recipients/consumers, which include:

- extended functionality including hygiene (a benefit for end users);
- the possibility of recycling using generally available methods (a benefit for the target group);
- pro-ecological solutions, namely a biodegradable product produced from renewable raw materials of natural origin (a benefit for the target group);
- a solution with increased innovation that is the same price as alternative solutions (a benefit for the target group); and
- reduced consumption of raw material.

This paper discusses studies that were carried out as part of a project whose aim was to develop an innovative line of functional paper towels containing strengthening additives obtained from recycled waste and functionalising biopolymer additives that replace water-fixing resins and carry innovative hygienising substances. The innovative functional paper towels developed were enriched with reinforcing additives in the form of nano/micro cellulose fibres obtained from biotechnological treatment of cellulose waste from the production of hygienic paper using specialised enzymatic preparations and functionalising additives based on biopolymers, including chitosan and starch as a carrier of Gemini surfactants. The novel functional paper towels were assumed to have three features compared with other solutions with a similar purpose. First, they have antimicrobial properties, including the ability to inhibit the growth of *Staphylococcus aureus*, *Bacillus subtilis* and *Escherichia coli* over time. This ability would reduce the possibility transmitting microorganisms and allowing them to multiply in workplaces, restaurants, public places, shopping centres, etc., by improving the effectiveness of removing microorganisms when drying hands after washing them (increasing the level of hygiene). Second, they allow the possibility of recycling while maintaining appropriate durability parameters of the product. This ability would reduce the amount of waste in landfills, allow the reuse of materials and raw materials or their processing into new ones, lower energy consumption and expenses and thus reduce the overall burden on natural environments. Finally, they show > 90% biodegradability over a period of 10 days to 8 weeks with over 99% content of biodegradable raw materials. This ability offers a fully environmentally friendly product.

Based on previous experience of the Łukasiewicz Łódź Institute of Technology (LIT) regarding water-fixing paper, chitosan was selected as one of the components of the polymer compositions. This cationic copolymer is composed of glucosamine and *N*-acetyl-glucosamine residues connected by a β -1,4-glycosidic bond. Chitosan has a reactive amino group at C2 and primary and secondary hydroxyl groups present at C3 and C6, thanks to which it has good adsorption properties [1, 2]. The use of chitosan should contribute to the elimination or a significant reduction in the use of resins, mainly polyamide-epichlorohydrin, and thus facilitate the pulping of paper products in the recycling process in an aqueous environment. The choice of chitosan for water-fixing paper was primarily based on its good miscibility with cellulose and a molecular structure that is similar to cellulose, which favours the formation of strong hydrogen bonds and thus increases the mechanical strength of the paper. An important biological property of chitosan is its antibacterial and antifungal activity [3, 4] from its positively charged amino groups, which react with negatively charged lipopolysaccharides and proteins on the surface of microbial cells, leading to disintegration of cell membranes and damage to the bacterial cell wall. The antibacterial effect of chitosan also varies depending on its degree

of deacetylation and molecular weight. High-molecular-weight chitosan can create a polymer film on the surface of the microbial cell wall to prevent the delivery of nutrients and thus causes the death of bacterial cells. However, low-molecular-weight chitosan can penetrate cells and combine with negatively charged intracellular components [5, 6]. A carrier of innovative hygiene substances (Gemini surfactants) developed based on the selected form of chitosan in the form of polymer-biocide mixtures should demonstrate increased antimicrobial activity. Gemini are highly effective against bacteria and microscopic fungi [7, 8]. Introducing both monomeric didecyltrimethylammonium chloride (DDAC) and dimeric hexamethylene-1,6-bis-(*N,N*-dimethyl-*N*-dodecylammonium bromide)C6 surfactants onto the surface of the paper via coating and spraying would provide good protection against microorganisms.

2. Materials and Methods

2.1. Materials

The following chemicals were used in the experiments: chitosan characterised by a degree of deacetylation of $\geq 90\%$, solubility of $\geq 99\%$ (in 1% acetic acid), a water content of $\leq 10\%$, an ash content of $\leq 1\%$, a protein content that is not detectable, a viscosity of < 200 mPa·s, a heavy metal content of < 10 ppm (Chemsta, Poland); the Gemini surfactant GEMSUR 12.06 and the cationic surfactant dibromide hexamethylene-1,6-bis-(*N,N*-dimethyl-*N*-dodecylammonium) (MDA, Poland); and starch HI-CAT 3353A (Roquette Group, France).

2.2. Methods

2.2.1. Preparation of Reinforcing Additives at the Laboratory Scale

Laboratory tests on the preparation of cellulose micro/nanofibers for use as reinforcing additives employed post-production waste cellulose materials obtained in the finishing stage of the production of paper towels and toilet paper. Before the enzymatic treatment process, cellulose waste was subjected to preliminary mechanical treatment to modify the structure of the initial waste masses. During this process, the cellulose structure loosens due to internal and external fibrillation due to the breaking of hydrogen and interfibrillar bonds. The following optimal parameters for enzymatic treatment of waste cellulosic materials at the lab scale were determined: for the enzyme, 900 UCMC per gram cellulose; a 2.5% cellulose suspension; and a reaction time of 2 h [9]. Figure 1 shows a scanning electron micrograph of waste cellulosic material after the enzymatic treatment and homogenisation stages.

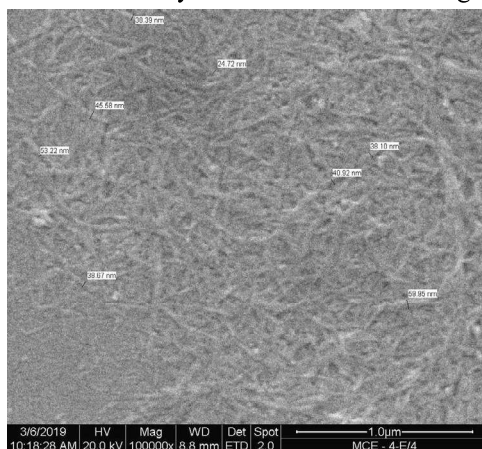


Figure 1. The scanning electron micrograph shows waste cellulosic material after the enzymatic treatment and homogenisation. The numbers indicate the dimensions of the fibres [9, 10].

2.2.2. Preparation of Reinforcing Additives at the Pilot Research Installation

Tests on the production of strengthening additives were carried out based on process assumptions developed under laboratory conditions. Grinding was carried out in a flow device with an actuating element design similar to the grinding head used during the development of the technology under laboratory conditions [9]. The pilot research installation comprised five stages [11].

- Stage I – defibration of production waste generated in the production of toilet paper in a centrifugal pulper. The process parameters were: water volume 700 dm³, paper weight 37 kg, concentration 5.5% and defibration time 15 min.
- Stage II – grinding the paper pulp obtained in Stage I using a centrifugal pulper, a pump transporting the pulp and a mill operating in a closed circuit with full circulation of the pulp. The process parameters were: grinding time 90 min and concentration 4%.
- Stage III – dilution and transport of mass to the enzymatic treatment reactor. The process was carried out using a pump transporting paper pulp and a water supply system. The process parameters were: input concentration 4% and final concentration 2.5%.
- Stage IV : enzymatic treatment of paper pulp in the reactor. The process parameters were: paper pulp concentration 2.5%, dry matter amount 37 kg, pulp volume 1480 dm³, amount of enzyme prepartate 0.89 dm³ and enzymatic treatment time 120 min.
- Stage V – dilution and homogenisation using a tank with a mixer and a homogeniser with continuous mixing. The process parameters were: flow through the homogeniser 200 dm³/h and fibre share in the composition 1.25%.

2.2.3. Preparation of Functionalising Additives in Real Conditions

The process to prepare functionalising compositions was developed based on guidelines developed during laboratory work [12]. It comprised four stages [11, 13].

- Stage I – dissolving chitosan in a lactic acid solution using a tank with a mixer, an industrial homogeniser and a circulation pump. The process parameters were: concentration of lactic acid solution 0.45%, amount of solution 80 dm³, chitosan share 2.4%, dissolution process time 20 min and deaeration time 15 min.
- Stage II – preparation of an aqueous starch solution using a tank with a mixer, an industrial homogeniser and a circulation pump. The process parameters were: water volume 80 dm³, starch content 1.2% and process time 20 min.
- Stage III – combining the chitosan solution with the starch solution using two tanks with mixers, a homogeniser and a circulation pump. The process parameters were: process time 20 min, final volume 160 dm³, final percentage of chitosan 1.2% and final percentage of starch 0.6%.
- Stage IV – introducing the Gemini surfactant into the composition using a tank with a mixer, homogeniser and circulation pump. The process parameters were: Gemini surfactant volume 1 dm³, Gemini surfactant concentration 0.6%, final volume 161 dm³, final percentage of chitosan 1.2% and final percentage of starch 0.6%.

2.2.4. Applying Separate Functionalising and Reinforcing Compositions to the Paper Web

Attempts to apply strengthening and functionalising compositions were made using a system of two separate spray collectors. The reinforcing additives were sprayed using

a system powered by peristaltic pumps and a system of rotary nozzles. The composition of functionalising additives was sprayed using flat-jet nozzles. The tests were carried out in the range of linear web speed from 100 to 150 m/min. The process parameters were: share of the composition of functionalising additives 60%, share of the composition of reinforcing additives 40%, spraying efficiency 144 dm³/h, efficiency of spraying functionalising additives 48–64 dm³/h and efficiency of spraying additives strengthening 52.8–74.4 dm³/h [11].

2.2.5. Analytical Methods

Antibacterial activity of the functional hygiene paper samples was assessed at the Accredited Laboratory of Biodegradation and Microbiological Research of Łukasiewicz – LIT. The antibacterial activity tests used *E. coli*, *S. aureus* and *B. subtilis*. Specifically, antibacterial activity against *E. coli* ATCC 11229 and *S. aureus* ATCC 6538 were carried out in accordance with PN-EN ISO 20743:2013 by counting on plates [14]. The criteria for assessing antibacterial activity are presented in Annex F of PN-EN ISO 20743:2013. The antibacterial activity against *B. subtilis* ATCC 6633 was tested using the parallel streak method in accordance AATCC Test Method 147:2011.

Biodegradation of the hygienic paper samples was assessed at the Accredited Laboratory of Biodegradation and Microbiological Research of Łukasiewicz – LIT. The tests were carried out in accordance with Procedure No. 2 (V Edition of 4 June 2018), ‘Determination of the degree of degradation of plastics and textile products in simulated composting conditions on a laboratory scale. Method for determining mass loss’ based on the following standards: PN-EN 14045:2012, PN-EN 14806:2010 and PN-EN ISO 20200:2016-01.

3. Results and Discussion

The technological tests regarding the production of functional hygienic papers were carried out in real conditions, including development related to optimisation of the scale to semi-technical and technical scale and integrating the application of reinforcing additives, and integrated reinforcing and functionalising compositions. The technological tests aimed at verifying the technological assumptions of the functional paper production process in real conditions using prototype modular devices integrated with a pilot research installation. Tests were carried out using a system with two separate spray collectors. The reinforcing additives were sprayed using a system powered by peristaltic pumps and a system of rotary nozzles. The functionalising additives were sprayed using flat-jet nozzles. Based on the results of previous tests, the only technological parameter that changed was the share of the active substance relative to the mass of the product. During the tests, different linear speeds of the paper ribbon were used to collect information that could to optimise the process technically and economically. The tests were carried out in the range of linear web speed from 100 to 150 m/min. During optimisation, a series of hygienic papers functionalised with biopolymer-biocide compositions based on chitosan, starch and Gemini surfactants were prepared, and the parameters for the production of papers functionalised with biopolymer compositions with expected functional properties were developed [12, 15, 16]. Based on previous experience, three paper samples were selected. Table 1 presents their technological parameters. The share of the active substance used, relative to the weight of the paper produced, was 0.5%, 0.6% and 0.69%. These samples were subjected to assays to assess antimicrobial activity and biodegradability.

Table 1. Technological parameters of hygienic papers modified with reinforcing and functionalising compositions.

Sample symbol	Additives [%]	Nozzle performance [dm ³ /h]	Spraying efficiency [dm ³ /h]	Linear speed of paper web [m/min]	Thickness of sprayed layer [mm]	Percentage of composition [%]	Active substance in composition [%]	Active substance in paper [%]
K2/1	40 Reinforcing	22	52.8	150	0.002560	8.26	6.00	0.50
	60 Functional	12	48.0		0.002608			
K2/2	40 Reinforcing	26	62.4	125	0.003438	11.09	5.40	0.60
	60 Functional	13	52.0		0.002287			
K2/3	40 Reinforcing	31	74.4	100	0.004099	13.22	5.20	0.69
	60 Functional	15	60.0		0.001711			

3.1. Antibacterial Activity Against *S. aureus* and *E. coli*

A specific number of bacteria was applied to the test and control samples. After incubation for 24 h, the change in the number of bacteria on the test and control samples was assessed, and the antibacterial effect was calculated. There were three replicates for each control (paper towel without additives) and test sample (0.4 ± 0.05 g) for each incubation time. The prepared bacterial suspension with a density of 10⁵ cells/mL was inoculated into previously sterilised control and test samples (0.2 ml per 0.4 g of sample; six replicates for each test sample and six replicates for the control sample). Three replicates of each sample were washed immediately, and three replicates of each sample were incubated at 37 ± 1°C for 24 h. After incubation, the bacteria were washed off by shaking in a neutralising Soya Casein Digest Lecithin Polysorbate (SCDLP) Broth solution. Appropriate dilutions were made from the obtained suspension in saline containing peptone. Each dilution was sub-cultured onto Plate Count Agar. The antimicrobial activity of the hygienic papers against *S. aureus* ATCC 6538 and *E. coli* ATCC 11229 are presented in Tables 2–4. The hygienic papers exhibited strong antibacterial activity against both strains.

3.2. Antibacterial Activity Against *B. subtilis*

The test samples were placed linearly on test plates with agar medium inoculated with a bacterial suspension. A paper towel sample with an active agent was used as an active control. A sample without antibacterial additive was used as an inactive control. The samples were incubated at 37°C for 24 h. After incubation, bacterial growth was assessed. A lack of growth under the sample and along the sides of the sample indicates that the sample has a bacteriostatic effect. A paper towel sample with an active agent was used as an active control. A sample without antibacterial additive was used as an inactive

Table 2. (continued) Assessment of the antibacterial activity of the hygienic paper samples against *Staphylococcus aureus* ATCC 6538.

Evaluated parameter	Requirement	Test results				Study evaluation
Sample K2/2						
Extreme difference in the logarithm from the number of bacteria on the test sample at time 0 and after incubation ($\log T_{\max} - \log T_{\min}$)	$\log < 2$	$\log T_{0\min} = 1.3$	0.0	$\log T_{T\min} = 1.3$	0.0	Fulfils condition
		$\log T_{0\max} = 1.3$		$\log T_{T\max} = 1.3$		
Growth value for the test sample	$\log T_t - \log T_0 = G$	0.0				Fulfils condition
Sample K2/3						
Extreme difference in the logarithm from the number of bacteria on the test sample at time 0 and after incubation ($\log T_{\max} - \log T_{\min}$)	$\log < 2$	$\log T_{0\min} = 1.3$	0.0	$\log T_{T\min} = 1.3$	0.0	Fulfils condition
		$\log T_{0\max} = 1.3$		$\log T_{T\max} = 1.3$		
Growth value for the test sample	$\log T_t - \log T_0 = G$	0.0				Fulfils condition

Table 3. Assessment of the antibacterial activity of the hygienic paper samples against *Escherichia coli* ATCC 11229.

Evaluated parameter	Requirement	Test results	Study evaluation											
Control sample (without additives)														
Inoculum concentration	$1-3 \times 10^5$ [CFU/ml]	2.6×10^5 [CFU/ml]	Fulfils condition											
Extreme difference in the logarithm of the number of bacteria on the control sample at time 0 and after incubation ($\log C_{\max} - \log C_{\min}$)	$\log < 1$	<table border="1"> <tr> <th colspan="2">Contact time 0 h</th> <th rowspan="2">Contact time 18–24 h</th> </tr> <tr> <td>$\log C_{0\min} = 4.03$</td> <td>0.04</td> </tr> <tr> <td>$\log C_{0\max} = 4.07$</td> <td></td> <td> <table border="1"> <tr> <td>$\log C_{T\min} = 5.99$</td> <td rowspan="2">0.23</td> </tr> <tr> <td>$\log C_{T\max} = 6.22$</td> </tr> </table> </td> </tr> </table>	Contact time 0 h		Contact time 18–24 h	$\log C_{0\min} = 4.03$	0.04	$\log C_{0\max} = 4.07$		<table border="1"> <tr> <td>$\log C_{T\min} = 5.99$</td> <td rowspan="2">0.23</td> </tr> <tr> <td>$\log C_{T\max} = 6.22$</td> </tr> </table>	$\log C_{T\min} = 5.99$	0.23	$\log C_{T\max} = 6.22$	-
		Contact time 0 h		Contact time 18–24 h										
$\log C_{0\min} = 4.03$	0.04													
$\log C_{0\max} = 4.07$		<table border="1"> <tr> <td>$\log C_{T\min} = 5.99$</td> <td rowspan="2">0.23</td> </tr> <tr> <td>$\log C_{T\max} = 6.22$</td> </tr> </table>	$\log C_{T\min} = 5.99$	0.23	$\log C_{T\max} = 6.22$									
$\log C_{T\min} = 5.99$	0.23													
$\log C_{T\max} = 6.22$														
Growth value for the control sample	$\log C_t - \log C_0 = F \geq 1$	2.02	Fulfils condition											
Sample K2/1														
Extreme difference in the logarithm from the number of bacteria on the test sample at time 0 and after incubation ($\log T_{\max} - \log T_{\min}$)	$\log < 2$	<table border="1"> <tr> <th colspan="2">0.0</th> <th rowspan="2">0.0</th> </tr> <tr> <td>$\log T_{0\min} = 1.3$</td> <td></td> </tr> <tr> <td>$\log T_{0\max} = 1.3$</td> <td></td> <td> <table border="1"> <tr> <td>$\log T_{T\min} = 1.3$</td> <td rowspan="2">0.0</td> </tr> <tr> <td>$\log T_{T\max} = 1.3$</td> </tr> </table> </td> </tr> </table>	0.0		0.0	$\log T_{0\min} = 1.3$		$\log T_{0\max} = 1.3$		<table border="1"> <tr> <td>$\log T_{T\min} = 1.3$</td> <td rowspan="2">0.0</td> </tr> <tr> <td>$\log T_{T\max} = 1.3$</td> </tr> </table>	$\log T_{T\min} = 1.3$	0.0	$\log T_{T\max} = 1.3$	Fulfils condition
		0.0		0.0										
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$\log T_{0\max} = 1.3$		<table border="1"> <tr> <td>$\log T_{T\min} = 1.3$</td> <td rowspan="2">0.0</td> </tr> <tr> <td>$\log T_{T\max} = 1.3$</td> </tr> </table>	$\log T_{T\min} = 1.3$	0.0	$\log T_{T\max} = 1.3$									
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$\log T_{T\max} = 1.3$														
Growth value for the test sample	$\log T_t - \log T_0 = G$	0.0	Fulfils condition											

Table 3. (continued) Assessment of the antibacterial activity of the hygienic paper samples against *Escherichia coli* ATCC 11229.

Evaluated parameter	Requirement	Test results				Study evaluation
Sample K2/2						
Extreme difference in the logarithm from the number of bacteria on the test sample at time 0 and after incubation ($\log T_{\max} - \log T_{\min}$)	$\log < 2$	$\log T_{0\min} = 1.3$	0.0	$\log T_{T\min} = 1.3$	0.0	Fulfils condition
		$\log T_{0\max} = 1.3$		$\log T_{T\max} = 1.3$		
Growth value for the test sample	$\log T_t - \log T_0 = G$	0.0				Fulfils condition
Sample K2/3						
Extreme difference in the logarithm from the number of bacteria on the test sample at time 0 and after incubation ($\log T_{\max} - \log T_{\min}$)	$\log < 2$	$\log T_{0\min} = 1.3$	0.0	$\log T_{T\min} = 1.3$	0.0	Fulfils condition
		$\log T_{0\max} = 1.3$		$\log T_{T\max} = 1.3$		
Growth value for the test sample	$\log T_t - \log T_0 = G$	0.0				Fulfils condition

Table 4. Antibacterial activity of the hygienic paper samples against *Staphylococcus aureus* ATCC 6538 and *Escherichia coli* ATCC 11229.

Sample	Incubation time [h]	Number of bacteria [CFU/g]	Antibacterial activity of A*	Growth value
<i>Staphylococcus aureus</i> ATCC 6538				
Control	0	3.3×10^4	-	1.91
	24	2.6×10^6		
K2/1	0	<20	5.1	0.0
	24	<20		
K2/2	0	<20	5.1	0.0
	24	<20		
K2/3	0	<20	5.1	0.0
	24	<20		
<i>Escherichia coli</i> ATCC 11229				
Control	0	1.1×10^4	-	2.02
	24	1.2×10^6		
K2/1	0	<20	4.8	0.0
	24	<20		
K2/2	0	<20	4.8	0.0
	24	<20		
K2/3	0	<20	4.8	0.0
	24	<20		

* Effectiveness of antibacterial properties
 Significant
 Strong

Value of antibacterial activity
 $2 \leq A < 3$
 $A \geq 3$

Table 5. Antibacterial activity of the hygienic paper samples against *Bacillus subtilis* ATCC 6633.

Sample	Growth intensity under the sample	Zone of inhibition [mm]	Study evaluation	
Inactive control (tissue sample)	Good	No zone	No antibacterial effect	
Active control (sample of tissue paper with an active agent)	Lack of growth	Average of 8.3	Antibacterial effect	
	K2/1	Pattern	No growth	Good antibacterial effect
		Pattern	No growth	
		Pattern	No growth	
	K2/1	Dots	No growth	Good antibacterial effect
		Dots	No growth	
Dots		No growth		
K2/2	Pattern	No growth	Good antibacterial effect	
	Pattern	No growth		
	Pattern	No growth		
	Dots	No growth		
	Dots	No growth		
	Dots	No growth		
K2/3	Pattern	No growth	Good antibacterial effect	
	Pattern	No growth		
	Pattern	No growth		
	Dots	No growth		
	Dots	No growth		
	Dots	No growth		

Table 6. Susceptibility of the modified hygienic paper samples to biodegradation in compost.

	Replicate	Biodegradation time, week [days]				
		4 [28]	6 [42]	7 [49]	8 [56]	9 [63]
K2/1		Weight loss [%]				
	1	46.7	65.6	86.7	96.6	100
	2	21.6	63.1	82.3	78.9	100
	3	34.9	71.7	97.4	100	-
		Average sample weight loss after 8 weeks: 92% Final average sample weight loss after 9 weeks: 100%				
K2/2	Replicate	Biodegradation time, week [days]				
		1 [7]	4 [28]	8 [56]	12 [84]	16 [112]
		Weight loss [%]				
	1	5.04	45.0	84.3	79.0	87.0
2	3.49	74.6	75.0	86.5	88.5	
3	1.22	32.1	74.6	83.6	76.5	
		Average sample weight loss after 8 weeks: 78% Final average sample weight loss after 16 weeks: 84%				
K2/3	Replicate	Biodegradation time, week [days]				
		1 [7]	4 [28]	8 [56]	12 [84]	16 [112]
		Weight loss [%]				
	1	1.49	30.2	71.2	77.9	91.5
2	1.04	35.0	72.1	85.0	84.4	
3	1.75	30.9	79.2	79.6	82.9	
		Average sample weight loss after 8 weeks: 74% Final average sample weight loss after 16 weeks: 86%				

4. Conclusions

Under optimal conditions, the modified hygienic papers produced at a pilot research installation presented substantial antibacterial activity against *S. aureus* (activity = 5.1, growth value = 0.0) and *E. coli* (activity = 4.8, growth value = 0.0) and good antibacterial activity against *Bacillus subtilis* (zone of inhibition of bacterial growth = 4.7–5.4 mm). Moreover, in compost conditions, > 90% of the samples had degraded within 8 weeks. The possibility of recycling cellulose fibres from production waste as a substitute for water-setting resins in towel paper is important from the point of view of implementing the goals of a circular economy.

5. Acknowledgements

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