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Biodegradable Nonwoven of an Aliphatic-Aromatic Copolyester with an Active Cosmetic Layer

Abstract

Polymeric products enriched with natural additives e.g. hyaluronic acid, plant hydrolates or collagen may find a wide application avenue due to the ever growing demand for natural cosmetics. The ecology of cosmetics is equally important. Therefore, in the work presented a biodegradable nonwoven was prepared with natural components as an active layer for cosmetic application. Synthesied was the biodegradable aliphatic-aromatic copolyester: poly (1.4 butylene succinate-co-glutarate-co-adipate-co-terephtalate) suitable for the forming of spunbond nonwoven. The active layer was deposited by padding on basic nonwoven. Assessed were microbiological and mechanical properties as well as the in – compost decomposition of the product. Activity against S. aureus and E. coli was tested and microbiological purity estimated. The best serviceable composition of the active layer was selected based on the microbiological properties tested. It contains sodium hialuronate, plant hydrolate of the bitter orange (Neroli), and collagen. Thanks to good mechanical properties, nonwoven made of aliphatic-aromatic co-polyester may serve as a carrier of the cosmetic layer in applications like face masks.

Key words: *aliphatic-aromatic polymer, cosmetic nonwoven, mechanical properties, hyaluronic acid, collagen, plant-hydrolates, spun-bonded, microbiological activity, biodegradation, polyester.*

Introduction

Sales of nonwovens amounted to 29 bln USD worldwide in 2015. Expected is an annual growth of 7.5%, boosting the market to a level of 48 bln USD in 2022 [1]. The market is fairly diversified; nonwovens find their way to numerous application domains. They are popular in agriculture, industry, civil engineering, medicine and cosmetics. Ever growing is the use of polyester and polypropylene in the manufacture of nonwovens [2, 3].

Sales of cosmetics enjoy a steady growth; the present world market level is worth 445 bln USD [4]. In 2016 cosmetics valued at 16 mln PLN were sold in Poland, making the country the sixth biggest market in Europe. The market value of the country is estimated to reach 20 mln PLN in 2021, fueled by rising internal consumption and exports to European countries and China [5].

The favourable market position encourages producers to find novelty ecological products which would satisfy ever more demanding consumers. Preference is given to natural cosmetics that are safe for humans and the environment, without artificial preservatives like parabens or tiomersal. Challenging is the selection of a blend of active substances which would both exert a good care effect and ensure preservation of the product.

Hyaluronic acid belongs to the family of glycosaminoglycans (GAGs). It is a linear biopolymer built up of repeated disaccharide units (D-glucoronic acid and N-acetyl-D-glucosamine) linked by β-1,3 and β-1,4-glycoside bonds. It appears mostly as salt- sodium-hyaluronate, and in its configuration it is identical throughout animal organisms, from humans to bacteria. Hyaluronic acid is highly hygroscopic – its molecule absorbs up to 250 molecules of water; it has interesting viscoelastic properties and high osmotic activity; and it attracts various ions like Ca^{2+}. It is a biocompatible substance suitable for use in cosmetics [6]. In the human organism, most of its amount (over 50%) appears in the skin. The content of hyaluronic acid in the organism decreases with age. It was found that preparations of hyaluronic acid with a molar mass over 100 kDa perfectly moisten the skin by providing an occlusive layer which prevents the loss of water; penetration of the acid through an intact epidermis is limited. Sodium- and potassium hyaluronate are the main active components of moistening, protective and anti-ageing cosmetics. They are currently used in creams, beauty masks, tonics and conditioners for care of the face, neck, around the eyes, as well as in products for preventing cellulites and stretch marks [7, 8]. 2% is the highest concentration of hyaluronic acid or its salt permitted in cosmetic products [9].

Hydrolates are multicomponent blends of secondary plant metabolites obtained by steam distillation of whole plants or their parts, The process yields a product containing oil and water with dissolved volatile compounds. Hydrolates have a slightly acidic pH close to that of the skin, as well as valuable biological properties, a delicate and pleasant fragrance, and they provide care to the skin. They are much more delicate than ethereal oils and do not cause irritation, which altogether makes them fit for the care of very sensitive skin [10, 11]. Popular in cosmetics are hydrolates from the bitter orange (Neroli) and damascena rose. They bear compounds such as limonene, nerolidol, terpineol, terpinyl acetate, farnesol (Neroli hydrolate) or geraniol, citronellol, phenylethyl alcohol and nerol (rose hydrolate) [12]. The hydrolates exert antibacterial activity mainly against *Pseudomonas aeruginosa* of the *Enterobacteriaceae* family in the case of Neroli hydrolate, while the rose hydrolate fights off *Escherichia coli, Pseudomonas aeruginosa, Bacillus subtilis, Staphylococcus aureus, Chromobacterium violaceum* and *Erwinia carotovora*. The hydrolates also reveal therapeutic properties when used as anti-depressive and spasmolytic agents and as a tonic for skin cleansing [11, 12].

Composed of 19 various amino acids, collagen is the main constituent of connective tissue. The biggest amount of collagen appears in the skin, where it makes up about 70% of all kinds of proteins. Collagen is widely used in cosmetics, drugs and aesthetic medicine for its biological properties [13]. It is responsible for the elasticity and tension of the skin and its proper moistening [14]. It is often labelled the "protein of youth" since it plays a crucial role in the human organism by influencing its biological age, juvenile appearance, vitality, immunity and the rate of regeneration. A complete regeneration of collagen proceeds only in a young, healthy organism up to the age of 25. Thereafter, the decomposition of collagen fibres surpasses their generation, marking the starting point of ageing and health deficiency [15]. Collagen is, therefore, a popular and valuable constituent of dietary supplements and cosmetics to improve the condition of skin, nails and hair.

Not only new materials and technologies are sought in the domain of synthetic polymers and biodegradable resins, but also the modification of well known polymers is pursued to prepare resins with improved processing and performance properties. Aliphatic and aromatic-aliphatic polyesters are an important group amongst biodegradable polymers [16]. To improve physical properties while preserving susceptibility to biodegradation, aromatic components were built into the main aliphatic chain of polyesters, thus obtaining aliphatic-aromatic copolyesters [17, 18].

At the ŁUKASIEWICZ Research Network – Institute of Biopolymers and Chemical Fibres, a technology was elaborated for the preparation of a biodegradable aliphatic-aromatic copolyester by melt polycondensation. Dimethyl terephtalate (DMT) makes up the aromatic component, while the commercial product Uniestrol comprises the aliphatic portion. The latter is a blend of dimethyl esters of di-carboxylic acids: succinic, glutaric and adypic. Synthesis of the copolyester is described elsewhere [19], which is the Institute's proprietary knowhow patent [20 applied for]. Aliphatic-aromatic copolyester is primarily destined for the manufacture of meltblown and spunbond nonwovens for use in agriculture (plant protection) and for the casting of film for packaging applications.

Materials and methods

Raw materials

Dimethyl terephtalate (DMT), from Mogilevkhimvolokno Mogilev, Bielorus, the commercial product Uniestrol, from Organika Co. Poland, and 1,4 butanediol, from PHH Standard Co, Poland, were used in the synthesis of poly (1,4 butylene-succinate-co-glutarate-co-adipate-co terephtalate) (IBPE). Uniestrol is a blend of: 15-25% dimehyl succinate, 50% dimetyl glutarate and 25-45% dimethyl adipate. Tetra-butyl-ortho titanate, made by Sigma – Aldrich, Germany, was used as a catalyst and sodium carbonate, from Chempur Co., Poland, as a nucleation agent. The synthesis was made in a 30 l reactor, built by Fourné, Germany.

Synthesis of copolyesters

Copolymers were prepared by a two-step melt polycondensation method. Into a 30 l stainless reactor at 100 °C in a nitrogen atmosphere were introduced 4200 g of DMT, 5500 g of butylenediol (BD), 4600 g of Uniestrol and 2.5 g of $Ti(OBu)_4$ as a catalyst. The reactor was then warmed up to in the range of 165 °C to 225 °C under atmospheric pressure, and methanol was distilled off the reactor. Excess of 1,4-butanediol ensures a high yield (90%) of the first stage of the process and prevents the escaping of volatile components in the second stage, at low pressure. Subsequently, 5.5 g of $Ti(OC_4H_9)_4$ was added, the pressure slowly reduced to 0.54 HPa, and the reaction temperature raised to 240 °C. Excess of 1,4-butanediol was distilled during that time. The total time of the polycondensation process was 100-135 min. Molten polymer was extruded in the form of 2-3 mm thick strings and pelletised.

Spinning of nonwoven fabrics

Nonwovens were spun on a laboratory stand, constructed by Polmatex-Cenaro, Poland. Polymer IBPE in two lots in a total amount of 16 kg was used in the experiments. In the course of spinning the polymer was kept in a HR 50 granulate dryer, made by Piovan Co, Poland, at 60 °C, where the dew point was 36.9 °C. A spinneret with 210 holes of 0.3 and 0.9 mm capillary diameter was used in the process. The throughput of a single hole was 0.48 g/min. IBPE nonwovens were prepared with a surface density of 75 g/m^2.

Application of collagen on IBPE nonwoven

Supplied by Proteina Co. Poland, collagen variant II with a 700 µg/ml content of hydroxyproline was applied on the polymeric base by padding. Collagen concentration in the solution is estimated by the content of the characteristic amino acid – hydroxyproline. Samples of the IBPE nonwoven were padded with collagen solution and then left on glass to dry up at ambient temperature.

Application of plant hydrolates and hyaluronates (HA) on IBPE nonwoven

An active layer was applied on the IBPE nonwoven by padding. It consisted of a 1% aqueous solution of blended sodium hyaluronates with a molar mass of 1320 kDa (HHA) and 10 kDa (SHA), supplied by Making Cosmetics (USA), and with the addition of a 30% solution of plant hydrolate:
- Hydrolate of bitter orange (Neroli), Biochemia Urody Co. Poland
- Hydrolate of damascena rose, Biochemia Urody Co. Poland

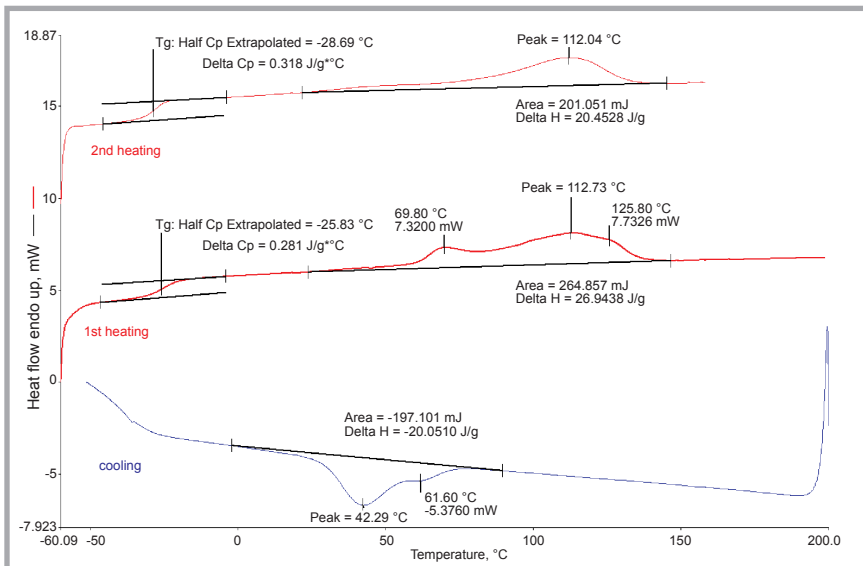The nonowoven was padded with the active solutions for 5 minutes and then put

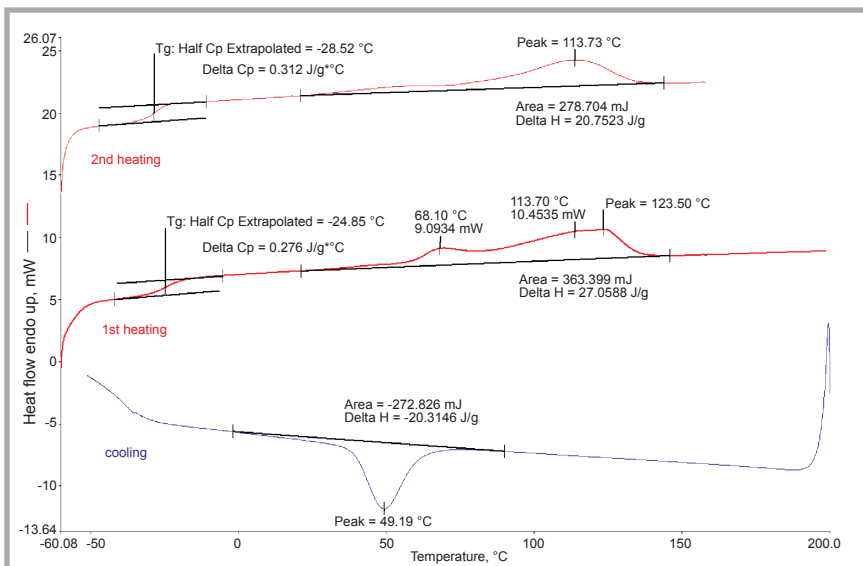*Figure 1. Thermogram DSC for polymer sample No 1.*

Figure 2. Thermogram DSC for polymer sample No 2.
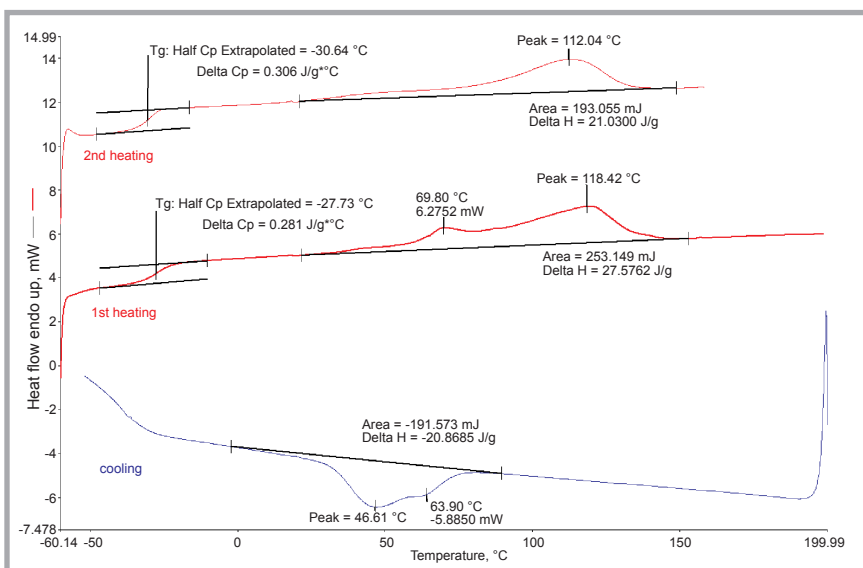

Figure 3. Thermogram DSC for polymer sample No 3.

onto teflon plates to dry out. The deposition degree of the active layer was estimated by the weighing method.

Materials

The following variants of the IBPE nonwovens were prepared by padding with various active components:

1) Nonwoven with a mixture of sodium hyaluronates and hydrolate of the damascena rose
2) Nonwoven with a mixture of sodium hyaluronates and hydrolate of the bitter orange (Neroli)
3) Nonwoven with collagen and a mixture of sodium hyaluronates and hydrolate of the bitter orange
4) Nonwoven with collagen and a mixture of sodium hyaluronates and hydrolate of the damascena rose
5) Nonwoven with collagen

■ Analytical methods

Assessment of physico-chemical properties of IBPE

Estimation of melt flow index, MFI

A molten sample at 190 °C was extruded through a spinneret with a 2.0 mm capillary, at a piston load of 2.16 kg. The extrusion proceeded in a defined time, and the MFI was calculated.

Estimation of relative viscosity

The relative viscosity (η_{rel}) of the IBPE was measured in its chloroform solution at 25 °C with a polymer concentration of 0.2 g/100 cm³. An Ubbelohde viscometer with a capillary 0A k = 0.00498 was used for the purpose.

DSC measurements

A thermal characteristic of the copolyester samples was prepared by differential scanning calorimetry with the use of Diamond, Perkin Elmer (USA) apparatus, equipped with an Introcooler II, microscale AD-2Z cooler for moderately low temperatures, and Pyris software.

Inspection of the structure

Composite materials were SEM-inspected by means of a microscope- Quanta 200 (FEI Co, USA) Samples powdered with a 20 nm layer of gold were tested in a high vacuum at an electron-beam-accelerating voltage of 5 KV.

Estimation of biodegradability [1]

Biodegradabiliy was tested in a compost medium by measuring the loss of

mass according to procedures PN-EN 14045:2005, PN-EN 14806:2010 & PN-EN ISO 20200:2007) [21-23].

The samples tested were treated with a compost medium containing aerobic microorganisms. Reactors with the samples being tested were placed in a thermal chamber in which constant conditions were maintained (temperature – 58 ± 2 °C, humidit – 40-70%). Mass loss was estimated by weighing after the incubation, which lasted from 1 to 24 weeks.

Assessment of microbiological activity [2)]

Antibacterial activity of the nonwovens prepared was assessed according to Standard PN-EN ISO 20743:2013 [24]. Bacteria strains from the American Collection of Pure Cultures (ATCC): *Staphylococcus aureus (*ATCC 6538) and *Escherichia coli* (ATCC 11229) were used in the tests. Samples of the nonwovens were inoculated with a bacteria suspension of defined concentration, and the number of bacteria was counted before and after the 24-hours incubation. A drop in the number in comparison to the reference samples before incubation marked the biological activity of the samples tested. The microbiological purity of the nonwoven was also estimated according to Polish Pharmacopeia [25].

Estimation of nonwoven mechanical properties [3)]

Tests were made at standard environmental conditions (20 ± 2 ºC and RH 65 ± 4%) according to Standard PN-EN ISO 139:2006 [26]. The following parameters were estimated:

- thickness – according to PN-EN ISO 9073-2:2002, with a thickness gauge TILMET – 64, constructed at the Lodz University of Technology, Poland. A pressure of 0.5 kPa [27] was applied;
- surface density – according to Standard PN-EN 29073-1:1994 [28];
- force and elongation – conditioned and wet according to Standard PN-EN 29073-3:1994. The measurements were made on a tenacity tester – Instron model 5544 (UK).

Distilled water at ambient temperature was used in the wet measurements of the virgin nonwoven, while in the tests with the remaining samples, a blend of active substances applied in the padding of the nonwovens was used [29].

- bending length along and cross – according to Standard PN-EN ISO 9073-7:2011 [30].
  The measurements were made on apparatus for measuring the flexural rigidity for samples cut along and across the nonwoven.
- ability to absorb liquid (LAC) and the capillary rise of the fluid – according to Standard PN-EN ISO 9073-6 [31]. Distilled water at room temperature was used as test liquid.

Deposition degree of the active substance (content of the active layer)

The deposition degree of the active substance was estimated by weighing with an accuracy of 0.0001g using a scale-Sartorius BP 110S (Germany).

■ Results and discussion

Synthesis of aliphatic-aromatic copolyester (IBPE)

IBPE belongs to the group of statistical copolymers with disordered regularity in their macromolecular chains and a decreased ability to crystallise. Sodiun carbonate decahydrate. in a suspension of 1,4 butanediol was therefore used as a nucleation agent. Three syntheses of the polymer were made at varied ester exchange times: 155 min (sample No 1), 100 min (sample No 2) and 135 min (sample No 3). Ester interchange was conducted at 240 °C to arrive at an adequate viscosity of the melt. Physical properties of the three polymer batches were analysed to select the one with the best parameters. Results are presented in *Table 1* and *Figures 1-3*.
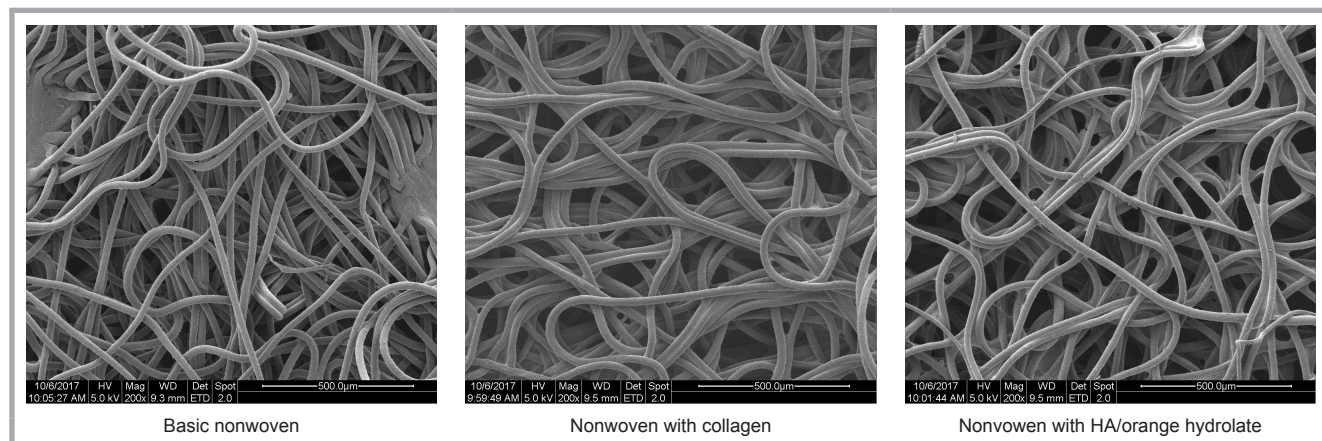
Table 1. Physical properties of IBPE.

Symbol No.	Ester exchange time, min	MFI, g/10 min	η relative, cm³/g	Tc, °C	ΔHc, J/g	Tg, °C	Tm, °C	ΔHm, J/g
1	155	22.0	115	42.3	-20.0	-28.7	112.0	20.5
2	100	29.7	114	49.2	-20.3	-28.5	113.7	20.7
3	135	42.7	113	46.6	-20.9	-30.6	112.4	21.0

Table 2. Content of the active layer.

Composition of the active layer	Average content, %
blend HA/ hydrolate of damascena rose	11.6
blend HA/ hydrolate of bitter orange	11.7
blend HA/ collagen/hydrolate of bitter orange	19.8
blend HA /collagen/hydrolate of damascene rose	19.7
Collagen	11.5

Figure 4. SEM images (magnification 200x) of IBPE nonwovens.

Basic nonwoven | Nonwoven with collagen | Nonvoven with HA/orange hydrolate
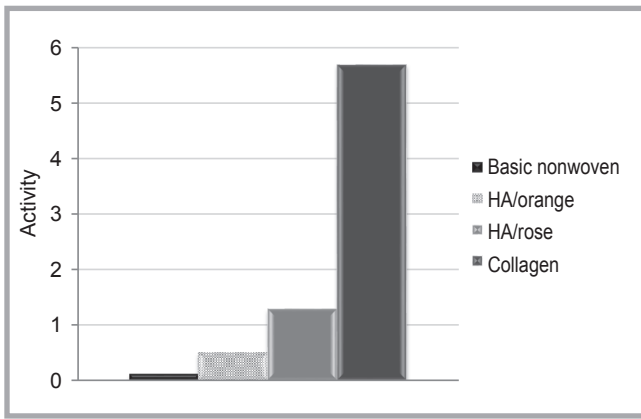
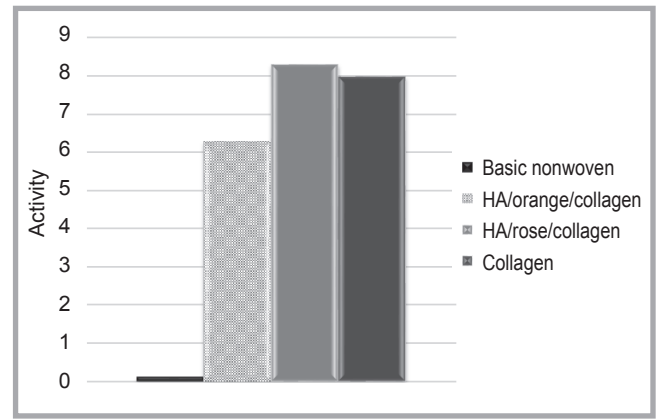**Figure 5.** *Activity of IBPE nonwovens against S. aureus.*

Figure 6. *Activity of IBPE nonwovens against E. coli.*

The IBPE samples show good thermal parameters, with the crystallisation temperature Tc being the most important – it should be beyond 40 °C to assure good performance of the polymer in the spunbond process. Samples 2 and 3 were selected for processing to nonwoven due to higher MFI values and better thermal properties. The higher the MFI (above 30 g/10 min), the easier the forming of the polymer into spunbond nonwovens runs.

Estimation of the content of the active layer

An active layer composed of collagen, a blend of sodium hyaluronates (HA),

and plant hydrolates was deposited onto the IBPE nonwoven by padding. The content of the layer in the singular nonwoven types was estimated by weighing. Results are shown in **Table 2.**

The contents of the active layers made up of sodium hyaluronates and different plant hydrates are close to each other, but it goes distinctly up with the addition of collagen. Generally, the contents are high, which is beneficial for cosmetic use.

Inspection of morphology

The basic nonwoven made of polymer IBPE, the nonwoven with collagen, and

the one with the HA blend and orange hydrate were inspected under a scanning microscope. An even distribution of the active layer on the fibres can be seen in **Figure 4**.

Microbiological assessment

Microbiological activity according to PN-EN ISO 20743:2013 [24] was estimated to assess the ability of the active cosmetic layer to reduce the bacteria number on the skin. It is a quantitative test for assessing the antibacterial activity of hydrophilic textiles after 24 hours of incubation. The examination was made with bacteria Gram (+) *S. aureus,* which is the cause of many dermatological infections [32]. Results for the IBPE nonwoven with deposited active layers are shown in **Figure 5**.

Nonwovens with a deposited active layer: one with HA and hydrolate of rose and the other with HA and hydrolate of bitter orange revealed activity against *S. aureus* of 1.28 and 0.48, respectively, signifying a lack of activity. Standard PN-EN ISO 20743:2013 quotes the range of 2 to 3 as substanial and above 3 as strong activity. The nonwoven with collagen showed 5.7, indicating highly effective action, which was probably caused by low pH (3.2-4.2) in collagen-variant II.

Figure 6 presents the activity of IBPE nonwovens with an active layer against bacteria Gram (–) *E. coli*.

The nonwovens with collagen, the HA blend and rose hydrate reveal the highest activity against *E. coli* at the level of 8.3. The nonwoven with collagen alone shows 8.0, while 6.3 is seen for the nonwoven with collagen, the HA blend and hydrolate of bitter orange. The addition of collagen made the materials acquire

Table 3. *Assessment of microbiological purity.*

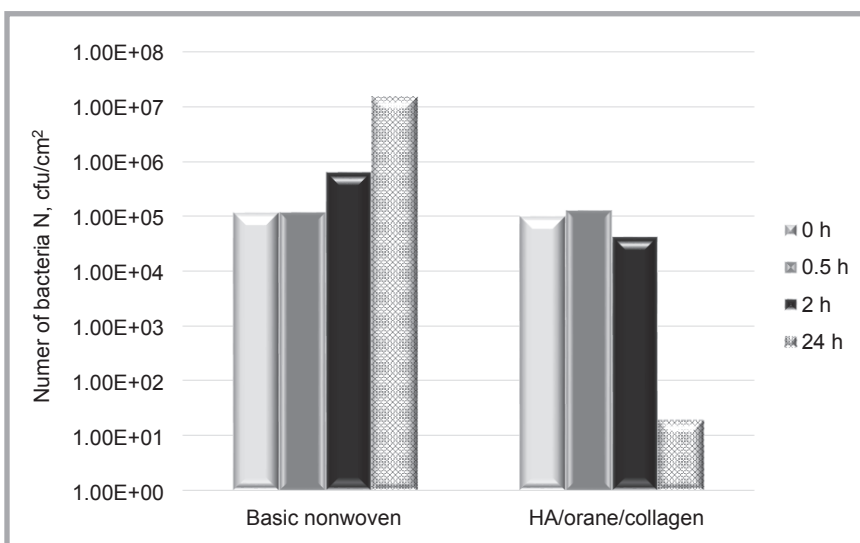| Nonwoven	Total number of mesophyllic bacteria, cfu/g	Total number of fungi, cfu/g	Limit from standard PN-EN ISO 17516:2014-11
Basic nonwoven	< 4.0 x 10¹	< 4.0 x 10¹	
Collagen	< 4.0 x 10¹	< 4.0 x 10¹	≤ 2.0 x 10³ jtk/g
HA/orange	2.4 x 10²	4.1 x 10¹	
HA/ rose/collagen	5.8 x 10¹	3.8 x 10¹	

Figure 7. *Comparison of time-dependent number of bacteria S. aureus.*

a strong antibacterial property against *E. coli* and *S. aureus* during 24 hours of contact. Such long contact of the nonwoven with the skin is not practicable in cosmetics, therefore the time of incubation for counting the number of *S. aureus* was, in further work, shortened for selected nonwovens. Results are presented in ***Figure 7***.

Right after incubation and after 30 minutes, the number of bacteria *S. aureus* on the basic nonwoven was constant at 1.2×10^5 cfu/ml; after 2 hours it began to rise (6.4×10^5 cfu/ml), and reached the level of 1.5×10^7 cfu/ml after 24 hours of incubation. The number of bacteria on the nonwoven with an active layer was slightly up at the beginning but went greatly down as soon as after 2 hours from the initial 1.5×10^7 cfu/ml; to 4.2×10^4 cfu/ml, and after 24 hours attained a level below 20 cfu/ml.

The nonwoven fabric with collagen, the HA mixture and bitter orange hydrolate cannot be use as an antibacterial agent on the skin due to the extended duration of action – above 2 hours. Nonwoven cosmetic products usually have a shorter time of contact with the skin: up to 0.5 hours for cosmetic masks and a few seconds for wet wipes. This feature, however, can be useful in protecting the product during prolonged storage and can act as a natural preservative.

Examined was also the microbiological purity according to Polish Pharmacopeia X [25], results of which are presented in ***Table 3***.

The basic nonwoven and those with the addition of collagen, the HA blend and hydrolates of bitter orange and damascena rose all satisfy the limit for cosmetic devices permitted, as quoted in Standard PN-EN ISO 17516:2014-11 [33].

Estimation of mechanical properties

Mechanical properties were tested for samples of basic nonwoven, nonwoven with a collagen layer, nonwoven with sodium hyaluronate, and nonwoven with hydrolate of bitter orange. Results are shown in ***Tables 4-9***.

It can be seen that the surface modification of the nonwoven with an active layer causes an increase in the breaking force and tenacity. Highest is the effect on the collagen-covered nonwoven Elongation

Table 4. Mechanical properties – conditioned.

Parameter	IBPE nonwovens		
	Basic nonwoven	HA/orange/collagen	Collagen
Thickness, mm	0.43	0.39	0.44
Surface density, g/m²	77.0	103.3	97.6
Breaking force along, N	3.34	4.04	6.02
Elongation along, %	90.8	50.9	49.0
Tenacity along, MPa	0.311	0.415	0.547
Breaking force across, N	2.33	2.81	4.16
Elongation across, %	101	63	96
Tenacity across, MPa	0.217	0.288	0.378

Table 5. Mechanical properties – wet.

Parameter	IBPE nonwovens		
	Basic nonwoven	HA/orange/collagen	Collagen
Thickness, mm	0.38	0.39	0.39
Breaking force along, N	1.94	1.53	1.49
Elongation along, %	52.7	43.0	52.0
Tenacity along, MPa	0.200	0.163	0.152
Breaking force across, N	1.56	1.19	1.29
Elongation across, %	79.7	60.4	76.5
Tenacity across, MPa	0.171	0.122	0.132

Table 6. Results of stiffness testing.

Parameter	IBPE nonwovens		
	Basic nonwoven	HA/orange/collagen	Collagen
Bending length along, cm	0.50	1.93	1.86
Specific bendig stiffness along, mN·cm	0.00788	0.7656	0.6467
Bending length across, cm	0.56	1.61	1.78
Specific bending stiffness, across, mN·cm	0.0137	0.4416	0.0822
General specific bending stiffness, mN·cm	0.0104	0.5814	0.6015

Table 7. Ability to absorb liquids – test results.

Nonwovens	Ability to absorb liquid LAC, %
Basic nonwoven	1.54
HA/orange/collagen	13.8
Collagen	13.9

Table 8. Capillary rise in lengthwise direction – test results.

Capillary rise along, mm		IBPE nonwovens		
		Basic nonwoven	HA/orange/collagen	Collagen
Time, sec	10	0	12.5	0
	30	0	20.5	0
	60	0	24.7	0
	300	0	36.0	0.6

Table 9. Capillary rise in widthwise direction – test results.

Capillary rise across, mm		IBPE nonwovens		
		Basic nonwoven	HA/orange/collagen	Collagen
Time, sec	10	0	16.5	0
	30	0	21.5	0
	60	0	26.2	1.6
	300	0	41.2	6.0

Table 10. *Results of biodecomposition tests.*

Sample	Time of biodegradation, week, day						
	1(7)	4(28)	8(56)	12(84)	16(112)	20(140)	24(168)
	Mass loss, %						
Polymer IBPE	0	1.77	10.1	17.7	25.5	30.3	41.1
	0	1.94	4.55	19.8	27.3	31.6	40.4
	0	2.16	6.01	16.3	25.1	31.5	39.1
Basic nonwoven	3.73	29.0	41.1	51.7	57.0	58.3	65.3
	2.88	24.4	44.3	54.9	64.1	64.1	64.1
	4.09	20.5	43.9	50.7	57.8	66.7	71.0
HA/ orange/collagen	18.6	53.3	54.6	71.2	72.0	73.6	77.5
	17.9	39.4	67.6	68.1	70.1	75.8	76.8
	16.8	33.6	53.0	61.8	67.2	67.8	77.7

both in the lengthwise and widthwise directions decreases in the covered nonwovens. This is explained by the higher stiffness conferred by the active additives.

It can be seen that the breaking force and tenacity are dramatically down in samples tested in a wet condition compared with the results of tests made with conditioned materials. It accords with the generally lower tenacity of wet synthetic fibres.

The aliphatic-aromatic polyester IBPE used has a larger aliphatic portion than the aromatic one (proportion Al/Ar is 60/40), which makes the aliphatic component undergo faster hydrolytic degradation under the influence of water. Polymers with a content of hetero-atoms (ester- and amide groups) in the main chain are more prone to hydrolytic rupture of the bonds [34,35]. Mechanical parameters of the nonwoven tested went down under the influence of the moisture-induced hydrolysis. Comparison of the breaking force and tenacity of the basic nonwoven with those of the covered nonwovens in a wet condition shows worse results for the latter, while elongation remains at a comparative level.

The nonwovens examined are characterised by a very low bending length, anticipating good drapeability. The bending length is up in nonwovens with the active layer applied, yet the drapeability at values below 2 cm (HA/orange/collagen) is still acceptable, pointing to the good performance of the products.

A very low absorption ability is shown by the basic nonwoven, but applying the active layer improves this. It may be inferred that collagen is the improving ingredient.

Capillary rise is one more parameter characterising the absorption ability of the nonwoven prepared. The basic nonwoven does not reveal any capillary effect, not even after 300 seconds. The use of the active layer HA/orange/collagen improves the capillary rise of the liquid, while a meager effect emerges with the collagen layer. The active layer composed of HA/orange/collagen substantially improves the absorption ability desired in the performance of the material.

Testing of biodecomposition

The examination of selected samples was made in aerobic compost in laboratory conditions. The biological activity and humidity of the inoculum used was estimated, where the total amount of the microorganisms was 5.1×10^7-1.1×10^8 jtk/g, and the humidity of the medium was in the range of 50.2% to 51.0%.

Table 10 presents results of the biodecomposition tests for the polymer, basic nonwoven and nonwoven with an active layer.

The starting polymer decomposes slowly, losing about 40% of its mass after 24 weeks. At the same time the mass loss of the basic nonwoven is down by about 65%; moreover a substantial mass loss (about 25%) can be seen as soon as after 4 weeks of the process. Furthermore, the addition of the active layer – collagen, hydrolate and HA insignificantly affects the speed of mass loss, which was about 77% after 24 weeks. However, a substantial loss (about 40%) was seen after 4 weeks of composting. The slowest degradation rate of the polymer results from its solid structural resistance to the penetration of microorganisms. The structure of the nonwoven is more

easily accessible, thereby promoting the biodecomposition. Since hyaluronic acid reveals a high affinity to water, nonwoven with its addition binds big amounts of water, providing conditions favourable for the growth of microorganisms as well as causing hydrolytic degradation.

◼ Conclusions

In this study, synthesized was biodegradable aliphatic-aromatic copolyester poly(1,4butylene-succinate-co-glutarate-co-adipate-co-terephtalate), named IBPE. The resin is characterised by good thermal properties, crystallisation temperature (T_c), in particular, and good rheology (Melt Flow Index) appropriate for the preparation of spunbond nonwovens.

The IBPE nonwoven prepared was of low tenacity and high elongation in both directions, and its surface was sufficiently even. Mechanical parameters of material both virgin and with an active layer applied were greatly down when measured in a wet condition. Mechanical parameters are not crucial in materials for cosmetic application. The nonwoven obtained was extendable, however, and not prone to breaking. Both the basic and covered nonwovens revealed good drapeability, favourable for use as a face mask. The basic nonwoven showed low fluid absorption of a level of 1.54%, since low fluid imbibition is inherently characteristic of polyester fibres. Absorption was distinctly up in 3 nonwovens covered with an active layer (high molecular hyaluronic acid has high affinity to water). All nonwovens with a collagen content revealed high antibacterial activity against *S. aureus* and *E. coli*. The antibacterial character of collagen – variant 2 probably stems from its low pH (3.2-4.2), which may inhibit bacteria growth and provide a peeling capability on face masks made of the material. The antibacterial action of collagen emerges only after more than 2 hours of incubation, and hence it is unsuitable as a bacteria-inhibiting agent in face masks. It may, however, be used as a skin conditioner and preserving agent.

Our investigation did not confirm literature information claiming the antibacterial property of plant hydrolates [11-13]. Different methods applied in estimating the property are the suspected reason for the divergent results of investigation. In this work hydrolates were applied on the nonwoven, thus deteriorating their activity.

All nonwovens examined satisfied the requirements for microbiological purity in cosmetic products as set out in Standard PN-EN ISO 17516:2014-11 [33].

An active layer consisting of collagen, orange hydrolate and a hyaluronic acid blend enhances the composting process because hydrolytic decomposition adds to the biodegradation process. Degradation products of the hydrolysis are more easily accessible for microorganisms, which makes the whole process run faster. Substantial mass loss could be seen as soon as after 4 weeks of composting, caused by the high affinity of HA to water molecules.

The biodegradable cosmetic nonwoven prepared with an active layer composed of collagen, a blend of sodium hyaluronates, and hydrolate of bitter orange satisfies the requirements for mechanical properties, and shows microbiological purity. The technology prepared enables preparation of a basic textile as a carrier for natural components in use for skin care.

Editorial notice

Acknowledgements

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