



Biodegradable self-adhesive tapes with starch carrier



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ABSTRACT

The disposed pressure-sensitive adhesive (PSA) tape widely used in daily life has been contaminating the environment and producing the vastly non-degradable trash. In this pioneering work, the advanced biodegradable pressure-sensitive double-coated tape containing starch carrier and water-soluble partially degradable modified pressure sensitive adhesive is architecturally designed and fabricated. The results have illustrated the excellent tack and peel adhesion of these newly constructed biodegradable self-adhesive tapes, and high thermal shear strength. Most importantly, the complete biodegradability of starch carrier and partial biodegradability of modified acrylic pressure-sensitive adhesives (PSA) have been confirmed experimentally. This environmentally friendly technology based on the starch resource utilization and novel water-soluble PSA will have great potentials for diverse applications such as the paper industry for manufacturing of ecological biodegradable product, the production of water-soluble biodegradable labels, medical tapes and biomedical electrodes.

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1. Introduction

Although biodegradable polymers have been commercialized for over 30 years, this niche market is beset with a variety of roadblocks led by high prices and lack of an industrial infrastructure to deal with these materials [1–3]. On the other hand, the wise utilization of bioresource is urgent regarding the emerging energy crisis and heavily environmental contaminations [4]. In fact, the biodegradable polymer can be derived from various raw materials such as chitosan, chitin and starch. Naturally, starch as the biopolymer in the living plants functions as energy storage material which is composed of amylopectin, amylase and glucose. As one of the most abundant biopolymers, starch can be smartly utilized as the low-cost and flexible component to architecturally fabricate composite materials for various applications [5–8]. Starch-based polymer composites are manufactured for diverse applications which are connected with excellent biodegradation under controlled conditions. The biodegradation of starch-based composites is determined by CO₂ and H₂O production and O₂ consumption by simultaneous changes in physical and mechanical properties of polymers containing unmodified or modified starch. Biodegradation of starch containing composites differently

proceeds under special conditions due to the diversity of the active microorganisms responsible for the degradation [9,10]. It is important to investigate the biodegradability of starch from the point of views of microbiology as well as the suitable microorganisms to obtain reliable conclusions. The chains in starch could also be broken down by non-enzymatic processes such as chemical hydrolysis [11,12]. Biodegradable polymers are often derived from plant consuming the atmospheric CO₂. Biodegradation process typically converts bioresources into CO₂, CH₄, water, biomass, humid matter, and other natural substances. Thus, they are naturally recycled by biological processes. Biodegradable polymers disposed in bioactive environments can be degraded by the enzymatic action of microorganisms such as bacteria, fungi, and algae [13,14]. Therefore, the starch-containing materials with the advanced properties and biodegradability are promising for the environmentally friendly applications [15,16].

Pressure-sensitive adhesives (PSAs) based on acrylics have been widely used in modern society [17–21]. The acrylic polymers in PSAs can persist for many years after disposal, and are not biodegradable. Built for the long haul, these polymers seem inappropriate for applications in which polymers are used only for a short-time period. Modified acrylic pressure-sensitive adhesives developed from various water-soluble derivatives such as ethoxylated amines, plasticizers and neutralization agents can be partially biodegradable. In fact, water-soluble pressure sensitive adhesives (PSA) based on acrylic is still a speciality. Water-soluble pressure sensitive adhesives (PSA) based on acrylic should be

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entirely water-soluble even in their crosslinked state while providing excellent thermal resistance, sufficient moisture resistance and very good adhesion to paper and other typical surfaces [22,23]. However, if the partially degradable acrylic based PSAs are used with the conventional polymer carriers, the biodegradability will be significantly affected.

Targeting at the utilization of native starch from bioresource, for the first time, the composite composing of starch carrier and acrylic-based PSAs with the partial degradability has been architecturally built and the adhesive properties and biodegradability have been examined.

2. Materials and methods

2.1. Raw materials

Potato starch was a product of Nowamyl S.A. (Poland), monohydrate citric acid (p.a) and glycerol (pure) were the products of Chempur (Poland). The distilled water was used throughout the whole experiment. All industrially raw materials were used for the synthesis and modification of water-soluble pressure-sensitive adhesives (PSA). Acrylic acid and butyl acrylate were purchased from BASF and AIBN was supplied by Dupont company with the trademark of Vazo 64. The details of other raw materials including the trademark (producer), chemical structure and function were presented in Table 1. The acetone and ethyl acetate were used as polymerization media and the isopropyl alcohol was used as stabilizers.

2.2. Manufacturing of starch films

Potato starch (10 g), glycerol (3 g), monohydrate citric acid (1.2 g), and distilled water (200 g) were placed in glass reactor. The reaction system was stirred mechanically and heated at 90 °C for 0.5 h for complete gelation of starch. After that, 50 ml of hot liquid was cast onto plastic Petri dish and placed in an oven at 60 °C for 24 h. The prepared starch film (carrier) were peeled off and used for further tests.

2.3. Synthesis of water-soluble acrylic PSA and evaluation of viscosity and molecular weight

The synthesis of pristine acrylic PSA was performed by the mixture of 70 wt% ethyl acetate and 30 wt% acetone in the presence of 0.1 wt% radical initiator azobisisobutyronitrile (AIBN) with the certain polymer amount (50 wt% of the prepared

solution). The monomers mixture containing 70 wt% butyl acrylate and 30 wt% acrylic acid was dropped in 2 h (dosage time) to the boiled solvent mixture at about 68 °C and the polymerization reaction conducted with 6 h post-reaction time for residue monomers. To further improve the water-soluble property of acrylic PSAs, the modification was carried out. The blended PSAs were prepared by the mixture of the pristine PSAs, crosslinker, plasticizer, tackifier and the neutralizing agent as Table 1 listed. Then, the mixture was further modified with crosslinking agent zirconium acetylacetonate (ZrACA), polypropylene glycol (Pluriol 400), ethoxylated diamine Ethoduomeen T/13 and potassium hydroxide (KOH) to achieve completely water-soluble pressure-sensitive adhesives. The viscosity of the investigated solvent-borne acrylics pressure-sensitive adhesives was determined with a Rheomat RM 189 from Rheometric Scientific, with spindle no. 3 at 23 °C. The molecular weight studies were performed with a liquid chromatograph LaChrom system: RI Detector L-7490 and LaChrom UV Detector L-7400 from Merck Hitachi, equipped with a PLgel 106 Å column from Hewlett Packard.

2.4. Preparing of double-coated biodegradable tapes with starch films as carriers

Water-soluble modified acrylic PSAs were coated on silicon paper with 75 g/m² coat weight dried for 10 min at 110 °C in drying canal and after that double-side coated under pressure on the afore-mentioned starch films. Then, the nascent double-sided self-adhesive tapes with starch films as carriers were tested on tack, and peel adhesion on steel and diverse papers, shear strength on steel, water solubility at pH: 7, 9 and 11, and biodegradability.

2.5. Measurement of starch film properties

Total soluble matter (TSM) was defined as the weight percentage of dissolved matter after 24 h immersion of the dried 0.2 mm thick starch film (15 × 5 mm) in 50 ml distilled water (at 23 °C). Mechanical properties such as elasticity module were determined using a tensile tester (Instron 4026, Instron Corporation). After 24 h conditioning in room humidity (RH)=55% six specimens (10 × 100 mm strips) were cut. The initial grip separation and cross-head speed were 50 mm and 10 mm/min, respectively.

2.6. Measurement of properties of self-adhesive tapes with starch films as carriers

The resulted biodegradable self-adhesive tapes were investigated for typical PSA properties, such as tack, peel adhesion and

Table 1
Raw materials for synthesis and modification of water-soluble acrylic PSA.

Trade name (Producer)	Abbreviation and chemical structure	Function
Zirconium acetylacetonate (Wacker Chemie)	$\text{ZrACA: Zr} \left[\begin{array}{c} \text{O} - \text{C} \begin{array}{l} \diagup \text{CH}_3 \\ \diagdown \text{CH} \end{array} \\ \text{O} = \text{C} \begin{array}{l} \diagdown \text{CH}_3 \\ \diagup \text{CH} \end{array} \end{array} \right]_4$	Crosslinker (Wacker Chemie)
Ethoduomeen T/13 (Akzo Chemie)	$\text{N,N',N''-Polyoxy-ethylene(10)N-faty-1,3-diamine propane:}$ $\text{R} - \text{N} - (\text{CH}_2)_3 - \text{N} \begin{array}{l} \diagup (\text{C}_2\text{H}_4\text{O})_x\text{H} \\ \diagdown (\text{C}_2\text{H}_4\text{O})_y\text{H} \end{array}$ $ \quad \quad \quad $ $(\text{C}_2\text{H}_4\text{O})_z\text{H}$	Modifying agents (Akzo Chemie)
Pluriol E 400 (BASF)	$\text{PEG 400: HO} - \left[\begin{array}{c} \text{CH} - \text{CH}_2 - \text{O} \\ \\ \text{CH}_3 \end{array} \right]_n \text{H}$	Water-soluble plasticizer

shear strength by standard A.F.E.R.A. (Association des Fabricants Européens de Rubans Auto-Adhésifs) procedures. Exact details could be found in AFERA 4015 (tack), AFERA 4001 (peel adhesion), and AFERA 4012 (shear strength). The data is reported as the average of the range of numbers observed during the test. The tack method according to AFERA 4015 is relatively simple and may be carried out using common tensile strength test machines. A sample of PSA-coated material 1-inch (about 2.5 cm) wide and about 7-inch (about 17.5 cm) long is bonded to a vertical of a clean steel test plate at least 10 lineal cm in firm contact. The vertical steel test plate is clamped in the jaws of a Zwick/Roell tensile testing machine. The scale reading in Newton is recorded as the tape is peeled from the steel surface with a constant rate of 100 mm per minute. For 180° peel measurements (AFERA 4001) the results depend on the face stock material. A sample of PSA-coated material 1-inch (about 2.5 cm) wide and about 5-in (about 12.7 cm) long is bonded to a horizontal target substrate surface of a clean steel test plate at least 12.7 cm in firm contact. A 2 kg hard rubber roller is used to apply the strip. The free end of the coated strip is doubled back nearly touching itself so the angle of removal will be 180°. The free end is attached to the adhesion tester scale. The steel test plate is clamped in the jaws of a tensile testing machine, which is capable of moving the plate away from the scale at a constant rate of 300 mm per minute. The scale reading in Newton is recorded as the tape is peeled from the steel surface. The data is reported as the average of the range of numbers observed during the test. The given result is an arithmetic average from the available peel adhesion results. Because the peel adhesion of novel tape was dependent on humidity, the peel adhesion of biodegradable self-adhesive tapes as a function of air humidity between 20% and 80% was evaluated. According to AFERA 4012, at 20 °C and at 70 °C, it is measured in N required to pull a standard area of adhesive coated sheet material from a stainless steel test panel under stress of a load between 5 and 90 N (tested at 20 °C) and between 5 and 40 N (tested at 70 °C). Static shear strength is the adhesive's ability to maintain its position when shearing forces are applied. Each test is conducted on an adhesive-coated strip applied to a standard stainless steel panel in a manner such that a 1-inch × 1-inch (about 2.5 cm × 2.5 cm) portion of the strip is in fixed contact with the panel with one end of the strip being free. The steel panel with the coated strip attached is held in a rack such that the panel forms an angle of 178–180° with the free end tape extended; the latter then is loaded with a force of different hanging weight in g or kg applied from the free end of the test strip. The time elapsed for each coated film to separate from the test panel was 4 h.

Water solubility (dispersibility) test (relates to water-soluble pressure-sensitive adhesives) is carried out at three pH values (pH=7, pH=9 and pH=11) at 20 °C on a 1 m × 1 m linen drape which is applied with 1 cm × 10 cm samples of pressure-sensitive adhesive prepared and autoclaved.

Dynamic shear cohesion was measured using a 2.5 cm × 2.5 cm end portion of a 2.5 cm × 15 cm strip of taped, formed by coating a 25 μm thick layer of adhesive on 68 g/m² super calendared Kraft paper backing, is adhered to a bright annealed steel test panel and rolled down with two passes of a 2 kg roller. The details can be found in the Test Methods for Pressure-Sensitive Tapes (Test PSTC-7).

2.7. Biodegradability of designed tapes

The biodegradation studies of starch carrier were performed using aerobic natural environmental conditions test. To plastic containers disinfected with ethanol compost soil (pH of compost soil solution: 7.2, humidity: 48%) was placed. Starch film was cut into square samples (60 × 60 mm), dried at 50 °C for 24 h and disinfected with ethanol. Subsequently, the samples were placed

in a container and embedded in soil. The test was done in triplicate. The containers were incubated at a constant temperature of 30 °C for 6 weeks. For that time humidity level was controlled and adjusted to be constant.

The rate of biological decomposition of the adhesive tapes was determined via the biochemical oxygen demand by measuring the O₂ partial pressure. The latter process not only provides a measure of the rate of organic constituents oxidative degradation at the microbiological level, but also provides evidence of the kinetics of decomposition via the corresponding curve shape. The calculation factor used for the rate of degradation is the chemical oxygen demand (7.800 mg O₂/l) determined experimentally by the potassium dichromate method, which can be taken as a measure of the complete mineralization of the organic substance contained in the product.

3. Results and discussion

3.1. Basic properties of biodegradable self-adhesive tapes

The synthesized water-soluble acrylic pressure-sensitive adhesive was characterized by the main parameters of viscosity, molecular weight and polydispersity which have been summarized in Table 2. For the useful application, the basic properties of pressure-sensitive adhesives should meet the requirements to ensure the elaborate adhesion/cohesion balance. Tack of pressure-sensitive adhesives is the adhesive property related to bond formation. The tack of biodegradable self-adhesive tapes was tested on steel and diverse kind of papers, such as fax paper, newsprint paper, photographic paper and art paper. As shown in Fig. 1a, the tack of developed biodegradable tapes evaluated on steel and selected papers stay on the stable level between 10.6 N and 11.2 N. This imparts the great potential of applying this kind of biodegradable tapes in the industry. Peel adhesion is another crucial factor which is related to the force required to remove a coated flexible pressure-sensitive adhesive sheet material from a test panel measured at a specific angle and rate of removal, reflecting the cohesion property. According to Fig. 1b, the results of peel adhesion test based on commercially water-soluble splicing tapes (samples 1–4) reveals that the adhesion values within the same paper groups ranges from 6.5 N to 9.5 N for fax paper, from 10.2 N to 11.8 N for newsprint paper, from 6.5 N to 9.1 N for art paper and from 8 N to 10.1 N for photographic paper. This is probably dependent on paper moisture which can influence the adhesion to mentioned paper surfaces. Fig. 2 shows shear strength of biodegradable self-adhesive tapes tested on steel in the high temperature range of 20 °C to 70 °C. As supposed, the increase of temperature influences negatively the measured cohesion on steel. The initial shear strength of 90 N at 20 °C was reduced on 40 N at 70 °C. In comparison with typical commercially splicing tapes used in the paper industry, the cohesion of 40 N at 70 °C is as very high to judge and recommended this biodegradable tape for splicing of papers in high temperatures.

The time of the water-solubility of water-soluble modified acrylic PSA has been examined. The water-solubility time is very short with 3 min 25 s for pH 7, 2 min 54 s for pH 9 and 2 min 33 s for pH 11. The increase of pH value allows for increasing of partial neutralization of carboxylic groups into polymer chain and

Table 2
Main parameters of synthesized PSA.

Viscosity (Pa s)	M_w (Dalton)	M_n (Dalton)	$P_d = M_w/M_n$
12.1	656 000	236 000	2.78

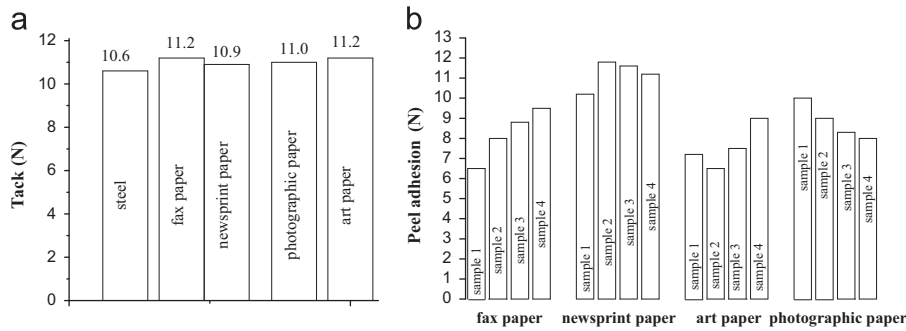


Fig. 1. Tack (a) and peel adhesion (b) of biodegrade self-adhesive tape on diverse substrate.

ameliorating the water-solubility. Fig. 3 shows the dynamic cohesion of typical commercially water-soluble splicing tapes to compare with a new generation biodegradable tapes based on water-soluble modified acrylic PSA and biodegradable starch carrier. The greatest reduction of dynamic cohesion was observed for sample 1. A little bit better results were shown for samples 2–4. The best thermal performance evaluated as dynamic cohesion as a function of temperature was observed for biodegradable tape put together from starch carrier coated double-sided with water-soluble modified acrylic PSA.

3.2. Biodegradability of novel self-adhesive tapes

Unmodified starch is biodegradable, low cost and renewable material [24]. Starch based foils prepared from native starch with glycerol as plasticizer were completely biodegradable after 35 days in the compost test [25]. In this work, three component systems: starch/glycerol/citric acid (citric acid worked as a crosslinking agent) was utilized. Solubility in water and Young module values of starch film were demonstrated in Table 3. After 42 days of soil test, all the samples were completely biodegraded (disintegrated). Additionally, the pH of soil solution was significantly reduced (6.06). This proves that the component system designed in this work has good biodegradability. The biochemical degradation of tape is approximately three-quarter finished after a period of twelve days as shown in Fig. 4. The end point of microbiological decomposition of the chemically oxidizable constituents is reached after approximately 23–25 days under the selected test conditions and is approx. 57%, which allows the adhesive tested to be classified as satisfactorily biodegradable. The water-soluble acrylic PSAs in this study are fully recyclable. They reached approx. 60% decomposition on the basis of the chemical oxygen demand (COD) of the test substance within 28 days in the BODIS test, provisional guideline of the German Federal Environmental Agency (UBA). It is therefore classified as partially biodegradable.

3.3. Peel adhesion of biodegradable tapes versus air humidity

The properties of tapes such as adhesion may depend on humidity. Therefore, the humidity dependence of adhesion is examined and the results are illustrated in Fig. 5. According to the obtained data, the dependence of adhesion on humidity can be determined from the following formula:

$$KK = A \cdot \left(1 - \frac{12}{10 \cdot RH}\right) \cdot \frac{(RH)^2}{200}$$

where KK is the adhesion at 20 °C with the unit of N, RH is the relative humidity (%) and A is the parameter dependence on the composition of water-soluble pressure-sensitive adhesive. Naturally, the peel adhesion of biodegradable tapes was influenced by the environmental air humidity. With increase of air humidity the

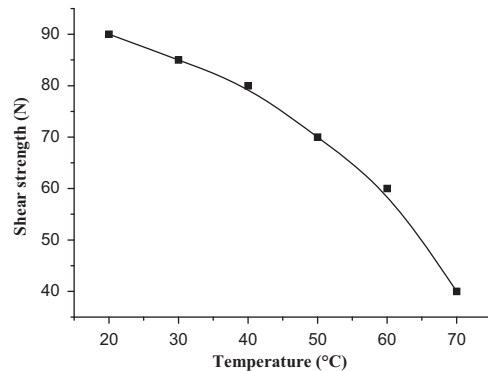


Fig. 2. Shear strength of biodegradable tape as a function of temperature.

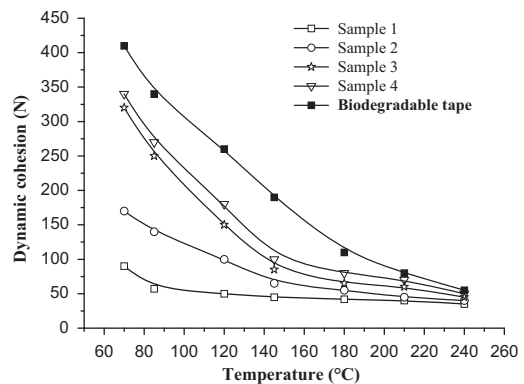


Fig. 3. Dynamic cohesion of tested water-soluble tapes.

Table 3 Properties of starch film.

Sample	TSM (%)	Young module (MPa)
Starch film	32	49

peel adhesion of biodegradable tapes increases. The water-soluble PSA layers and water-dispersible starch carrier are very highly hydrophilic and they can with it absorb the moisture which makes the PSA layers very soft.

4. Conclusions

The tack, adhesion and cohesion properties of novel biodegradable tape developed in this study are excellent. Compared with the

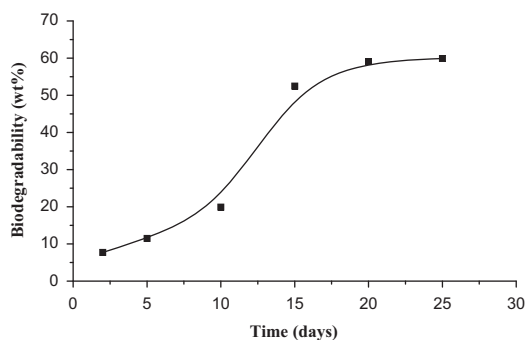


Fig. 4. Biodegradability process of water-soluble modified acrylic PSA.

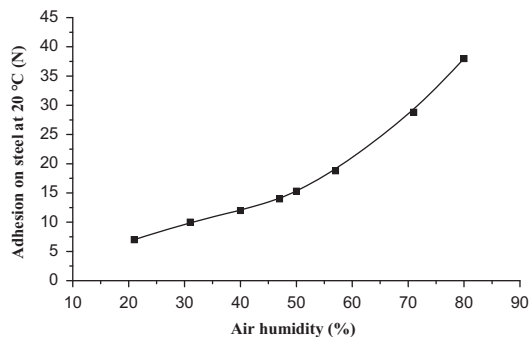


Fig. 5. Effect of air humidity level to adhesion of biodegradable tape on steel.

commercial tapes, the novel biodegradable tape with starch as carrier demonstrates the best thermal performance. After 42 days of soil test, all of starch-based carrier materials completely biodegrades. The water-soluble acrylic PSAs are fully recyclable, which reaches 60% decomposition on the basis of the chemical oxygen demand (COD) of the test substance within 28 days. The biochemical degradation of the developed tape is approximately three-quarter finished after a period of twelve days. This kind of products can potentially play a major role in the production of water-soluble biodegradable labels, medical tapes and biomedical electrodes, and similar products are still in the development stage.

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References

- [1] Pack S, Bobo E, Muir N, Yang K, Swaraj S, Ade H, et al. Engineering biodegradable polymer blends containing flame retardant-coated starch/nanoparticles. *Polymer* 2012;53:4787–99.
- [2] Liu WQ, Budtova T. Ionic liquid: a powerful solvent for homogeneous starch-cellulose mixing and making films with tuned morphology. *Polymer* 2012;53:5779–87.
- [3] Baji A, Wong SC, Liu TX, Li TC, Srivatsan TS. Morphological and X-ray diffraction studies of crystalline hydroxyapatite-reinforced polycaprolactone. *J Biomed Mater Res B* 2007;81B:343–50.
- [4] Wang JP, Chen YZ, Zhang SJ, Yu HQ. A chitosan-based flocculant prepared with gamma-irradiation-induced grafting. *Bioresour Technol* 2008;99:3397–402.
- [5] Pramoda KP, Mya KY, Lin TT, Lu XH, He CB. Investigation of thermomechanical properties and matrix-filler interaction on polyimide/graphene oxide composites. *Polym Eng Sci* 2012;52:2530–6.
- [6] Shao L, Bai YP, Huang X, Meng LH, Ma J. Fabrication and characterization of solution cast MWNTs/PEI nanocomposites. *J Appl Polym Sci* 2009;113:1879–86.
- [7] Gu HB, Huang YD, Zhang X, Wang Q, Zhu JH, Shao L, et al. Magneto-resistive polyaniline-magnetite nanocomposites with negative dielectrical properties. *Polymer* 2012;53:801–9.
- [8] Lau CH, Liu SL, Paul DR, Xia JZ, Jean YC, Chen HM, et al. Silica nanohybrid membranes with high CO₂ affinity for green hydrogen purification. *Adv Energy Mater* 2011;1:634–42.
- [9] Maiti S, Ray D, Mitra D, Sengupta S, Kar T. Structural changes of starch/polyvinyl alcohol biocomposite films reinforced with microcrystalline cellulose due to biodegradation in simulated aerobic compost environment. *J Appl Polym Sci* 2011;122:2503–11.
- [10] Lucas N, Bienaime C, Belloy C, Queneudec M, Silvestre F, Nava-Saucedo JE. Polymer biodegradation: mechanisms and estimation techniques. *Chemosphere* 2008;73:429–42.
- [11] Shi R, Zhu AC, Chen DF, Jiang XJ, Xu XC, Zhang LQ, et al. In vitro degradation of starch/PVA films and biocompatibility evaluation. *J Appl Polym Sci* 2010;115:346–57.
- [12] Pyla R, Kim TJ, Silva JL, Jung YS. Enhanced antimicrobial activity of starch-based film impregnated with thermally processed tannic acid, a strong antioxidant. *Int J Food Microbiol* 2010;137:154–60.
- [13] Avella M, De Vlieger JJ, Errico ME, Fischer S, Vacca P, Volpe MG. Biodegradable starch/clay nanocomposite films for food packaging applications. *Food Chem* 2005;93:467–74.
- [14] Doi Y. Biodegradable plastics and polymers. *J Pestic Sci* 1994;19:S11–4.
- [15] Khaili HPSA, Bhat AH, Yusra AFI. Green composites from sustainable cellulose nanofibrils: a review. *Carbohydr Polym* 2012;87:963–79.
- [16] Tondi G, Wieland S, Wimmer T, Schnabel T, Petutschnigg A. Starch-sugar synergy in wood adhesion science: basic studies and particleboard production. *Eur J Wood Wood Prod* 2012;70:271–8.
- [17] Czech Z, Kowalczyk A, Pelech R, Wrobel RJ, Shao L, Bai Y, et al. Using of carbon nanotubes and nano carbon black for electrical conductivity adjustment of pressure-sensitive adhesives. *Int J Adhes Adhes* 2012;36:20–4.
- [18] Jin X, Bai YP, Shao L, Yang BH, Tang YP. Properties of solvent-borne acrylic pressure-sensitive adhesives synthesized by a simple approach. *Express Polym Lett* 2009;3:814–20.
- [19] Wang WZ, Wang R, Zhang C, Lu S, Liu TX. Synthesis, characterization and self-assembly behavior in water as fluorescent sensors of cationic water-soluble conjugated polyfluorene-b-poly (N-isopropylacrylamide) diblock copolymers. *Polymer* 2009;50:1236–45.
- [20] Kong WS, Ju TJ, Park JH, Joo SR, Yoon HG, Lee JW. Synthesis and characterization of hydrogenated sorbic acid grafted dicyclopentadiene tackifier. *Int J Adhes Adhes* 2012;38:38–44.
- [21] Lee SW, Park JW, Kwon YE, Kim S, Kim HJ, Kim EA, et al. Optical properties and UV-curing behaviors of optically clear semi-interpenetrated structured acrylic pressure sensitive adhesives. *Int J Adhes Adhes* 2012;38:5–10.
- [22] Czech Z, Loclair H. Investigations of UV-crosslinkable water-soluble acrylic pressure-sensitive adhesives. *Polimery-W* 2005;50:64–8.
- [23] Czech Z. Acrylate-based water-dispersible or water-soluble pressure-sensitive adhesive tapes. *Polimery-W* 1996;41:22–30.
- [24] Lu DR, Xiao CM, Xu SJ. Starch-based completely biodegradable polymer materials. *Express Polym Lett* 2009;3:366–75.
- [25] Domka L, Malicka A, Jagla K, Kozak A. Biodegradation of starch-modified foil in natural conditions. *Pol J Environ Stud* 2009;18:191–5.