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Influence of ultrasonic treatment on properties of bio-based coated paper

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ABSTRACT

In the research, kraft paper was coated with a blend of chitosan and rice starch ultrasonic-treated coating solution. The novelty in this paper is the use of bio-based coating solution which was treated with ultrasound, which led to a positive effect on the mechanical, optical and water barrier properties. The goal of the research was to improve the coating solution with ultrasound and to make the coated kraft paper with chitosan and rice starch suitable for a packaging material. To evaluate the influence of the ultrasonic treatment on coating solutions, the moisture content, water vapour permeability, absorptiveness, tensile strength, elongation at break, tear and bursting strength, colour values, gloss and surface properties of coated paper were determined. The research showed that the moisture content in all coated papers decreased by less than 0.5%, the water vapour permeability reduction being around 20%. Tensile strength, elongation at break and tear strength improved at coated paper, even more at the one pretreated with ultrasound. Bursting strength, which is important at paper bags, increased by 9% at coated, and by 12% at coated and ultrasound-treated paper.

Furthermore, gloss improved at both coated papers. As expected, the coating affected the surface, with fewer and smaller pores. At paper the coating of which was treated with ultrasound the surface was smoother and fewer pores were detected. The results showed that the ultrasonic treatment is not only an inexpensive and environmentally friendly process, but is also very effective in the preparation of solutions for bio-based coatings. Such coatings have better moisture resistance and are more flexible, which is important for packaging paper. Therefore, preparing solutions using ultrasound as pretreatment is an improved and convenient procedure to enhance many properties of bio-based coatings.

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1. Introduction

In recent years, the materials used in the packaging of goods, such as fruit, vegetables, bakery products or flowers, have improved in developing recyclable packaging from waste or abundant materials. Products with low recyclability have a great environmental impact after its use and also during the production. Therefore, the manufacturers of bio-based materials are keen to replace oil-based packaging materials with more sustainable ones which also boast of improved barrier properties [1,2].

Kraft paper is paper with high elasticity and tear resistance, used for products which require good strength and durability [3]. Such paper can be used as packaging for food, consumer goods, flour bags, or even for cement. By coating kraft paper with bio-based polymers, such products have added value and improved certain

properties, depending on the used coatings. The purpose of the coating is to create conditions that a particular packaging needs. It is very important to choose the right coating, which will act as a binder on the surface of the paper and will not diminish the existing properties. One of the most commonly used natural coating polymers are polysaccharides (e.g. chitosan, chitin, starch, xylan etc) in all fields of materials science, also in packaging. Chitosan is one of the most non-toxic and commonly used polysaccharides, which is derived from the deacetylation of chitin [4]. Chitosan has also attracted interest in packaging, especially in food packaging as edible films and coatings [5,6]. Due to its good barrier properties (antimicrobial, mechanical, against grease, oxygen), chitosan coatings can be used as barriers in packaging. Chitosan has high crystallinity and a hydrogen bond between molecular chains, which exhibit great oxygen properties. Due to the positive charge on the amino group under acidic conditions, chitosan binds negatively charged molecules and therefore represents a greater barrier against grease [6].

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Previous research showed that chitosan exhibits good moisture properties, but not excellent, since it has a hydrophilic surface, which attracts moisture [7]. In order to improve the functional properties of chitosan coatings, a blend with other biopolymers, hydrophobic substances was proposed [8–10]. Numerous chitosan coatings and blends with other natural polymers were made [11,12]. Rice is the most widely used basic food in the world and due to different climates, soil characteristics and cultures, there are more than 240,000 registered varieties of rice and just as many different rice starches [13]. Due to the high amount of amylase, rice starch is attractive for food packaging as a film barrier. It has also been used to replace plastic film barriers as it has good mechanical properties [14–16]. From the literature, it is known that with an ultrasound treatment and with a combination of rice starch, the properties of composite films have improved [11]. The most important role of starch in the coating process is to act as a binder. On the other hand, it brings paper stiffness, which is important for runnability and printability. The use of starch in paper coatings should be careful, since in higher amounts, the surface elasticity is not sufficient and can therefore not be suitable for packaging products. Starch is sensitive to water and in many cases, in order to have an efficient coating, other polysaccharides are included into the coating mixture.

To improve the mechanical and moisture barrier properties of bio-based coatings, a blend solution of chitosan and rice starch was made, and the dispersion was before the coating treated with ultrasound.

Ultrasonic technology is environmentally friendly and is used in many fields, e.g. pharmaceutical, chemical industries etc. The use of this technology can lead to many improvements in materials. As it can be read in the literature, the positive effect of ultrasonic treatment on the gelatinisation of starch dispersions and the application of ultrasonic treatment to starch films improves the moisture properties of treated starches and provides stronger structure [17,18]. Previous research shows that Bourtoom & Chinnan studied the effect of rice starch, which was incorporated into chitosan film. The composite film showed an increase in tensile strength, moisture, water vapour permeability, whereas the elongation at break decreased [11].

The aim of the research was to improve the mechanical and moisture properties of bio-based coating on the packaging paper by using an ultrasonic treatment. The literature shows no record of previous research done on the blend of rice starch and chitosan coating improved with ultrasound. Our research was focused on the effect of ultrasonic treatment on the solutions for the preparation of the mentioned coating to investigate the possibility of producing bio-based coated paper for packaging and also to provide an effective water barrier when for example packaging flowers, not diminishing the existing properties. Using ultrasound is an environmentally friendly process and can be used for all solutions in order to improve biodegradable coatings. Such treatment and coating materials could be used as a substitute for the packaging coatings that are currently on the market.

2. Materials and methods

2.1. Materials

Chitosan, with the molecular weight lower than 20 kDa and deacetylation degree higher than 85%, was purchased from Sigma Aldrich (Austria). Malic acid (98%) was purchased from Sigma Aldrich (Austria). Rice starch was obtained from Farmalabor Srl (Italy) and had 14% of moisture content, 1% of proteins and 0.6% of ashes. Kraft paper (80 gm^{-2}) was provided by Papirnica Vevče, Slovenia.

2.2. Methods

2.2.1. Preparation of coating dispersions, using ultrasonic treatment

The rice starch solution was prepared by dissolving 4 g of rice starch in 100 ml of distilled water. The solution was mixed until it gelatinised (85°C for 5 min) and then cooled to 27°C . The chitosan solution was prepared by dissolving 2 g of chitosan in 100 ml of malic acid. The rice starch-chitosan solution was prepared by mixing 100 ml of 2% rice starch solution with 100 ml of 2% chitosan solution. After that, one solution was put into an ultrasonic bath (Asonic, Ultrasonic bath) for 15 min, using constant 35 kHz frequency. After the ultrasound treatment, the solutions were left in the bath for another 15 min.

After the mixed solution was treated, it was filtered through a polyester screen (mesh no. 140, mesh opening $160 \mu\text{m}$) with aspiration to remove small lumps in the solution, while the other, not treated, solution was filtered directly.

2.2.2. Coatings on kraft paper

After the solutions were prepared and treated with ultrasound, the coating with a coater proceeded. Kraft paper was coated with bio-based film forming solutions (untreated and treated with ultrasound) on a coating table at ambient temperature, using a $120 \mu\text{m}$ blade. Afterwards, it was dried at 80°C for 10 min. On each paper, 5 gm^{-2} of solution was applied. For a comparison, the uncoated as well as coated paper were analysed in the research.

2.2.3. Characterisation of samples

2.2.3.1. Grammage, thickness, density and specific surface volume. It was necessary to determine grammage to obtain the same amount of coating on all samples. Grammage was determined in accordance with the ISO 536 standard, 10 samples of each paper were cut into size $10 \times 10 \text{ cm}$ and weighed.

The thickness of samples was measured with a precision digital micrometre Mitutoyo Corporation, Japan, to the nearest $0.0001 \mu\text{m}$ at 10 random locations on each paper.

Density and specific surface volume were calculated from grammage and thickness, as described in the standard method ISO 534.

2.2.3.2. Moisture content, water absorption capacity (Cobb value) and water vapour barrier properties (WVP). Moisture content was determined by measuring weight loss after the drying in a laboratory oven at $105 \pm 1^\circ\text{C}$ until constant weight. Five samples of each paper were tested and the results were expressed in percentage. The moisture content was determined with ISO 287.

Water absorptiveness was determined with the Cobb value, as described in the standard method ISO 535, where a given amount of water was in contact with the paper for 60 s and weight differences were compared. For each paper, ten sample tests were made.

To determine the water vapour permeability (WVP) of uncoated and coated paper, the ISO 2528 standard method was used. Test cups were filled with water. A sample was placed between the cup and the ring cover of each cup. To ensure the best results of WVP, a silicone sealant was applied around the cup edge. Papers with the exposed area of 50 cm^2 were tested at $90 \pm 2\% \text{ RH}$ and $38 \pm 2^\circ\text{C}$ for 24 h. WVP was expressed in gram units per square meter per day. Three samples per each sample paper were tested.

2.2.3.3. Mechanical properties. Tensile strength (TS) and elongation at break (E) of papers were determined on a tensile testing machine Instron 6022. The samples were analysed in the standard atmosphere at 23°C of temperature and 50% of relative humidity. The cross speed head was 0.15 mm/s . Paper stripes of 18 cm in length and 1.5 cm in width were used and a minimum of ten probes for each sample in the machine direction (MC) and cross machine

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