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PREPARATION AND PROPERTIES OF AGGLOMERATED CORK PANELS BOUND WITH CHITOSAN BINDER

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ABSTRACT

In this paper, acidified chitosan was used as an adhesive to prepare aldehyde-free, environmentally-friendly agglomerated cork panels by hot-pressing. After preparation, the physical, mechanical, and the finishing properties of the chitosan-glued agglomerated cork panels were investigated. The optimal mass ratio of acetic acid solution (1 wt.%) to chitosan was determined to be 30:1. The resulting hot-pressed agglomerated cork panels, which featured a density of 0.55 $g \cdot cm^{-3}$ and a thickness of 4 mm, exhibited a tensile strength of 1.70 MPa and a thermal conductivity of 0.11 $W \cdot m^{-1} \cdot K^{-1}$. The agglomerated cork panels coated with the oil-based polyurethane and water-based, acrylic-modified polyurethane paints exhibited significantly lower lightness and higher glossiness. The total color differences (ΔE^*) of both agglomerated cork panels increased before and after finishing. The oil-based polyurethane paint coating exhibited high adhesion of paint film, reaching a level-0 adhesion, while the water-based, acrylic-modified polyurethane paint coating achieved a level-1 adhesion. The abrasion resistance results showed that the substrates of cork agglomerates coated two types of paint did not expose after undergoing abrasion for 100 revolutions at the turntable speed of 60 rpm.

KEYWORDS: Chitosan adhesive, agglomerated cork, mechanical properties, painting properties.

INTRODUCTION

Cork refers to the outer tissue layer of bark harvested from the trunk or branches of the *Quercus variabilis* or *Quercus suber* oak trees. Anatomically, cork belongs to the periderm (Leite and Helena 2017, Song and Zhao 2017). It has many useful properties, i.e. light weight, chemical stability, hydrophobicity, high elastic compression and dimensional recovery, and so on (Gil 2014, Pereira 2013). *Quercus suber* is mainly grown in European and Northern African countries, such as Portugal, Spain, Algeria, and Tunisia (Mateus et al. 2017). Cork derived from *Quercus suber* is mainly used commercially as wine stoppers. The poor-quality cork or residue from cork stoppers processing is processed into various particles for a variety of different application, such as insulation materials, decoration materials, cork mats, and various accessories, to name a few (Branco et al. 2021, Knapic et al. 2016, Aroso et al. 2017). In China, cork is harvested from the bark of *Quercus variabilis* trees, which grow slowly, and the cork tends to have a higher bulk density, higher hardness, higher impurity content, and lower quality compared to the cork from *Quercus variabilis*; the cork in China is mainly processed into cork particles to produce cork agglomerates by adding adhesives, leading to expanding the use of cork (Song et al. 2017, Wu et al. 2018, Zhao et al. 2013).

Industrial cork agglomerates often use polyurethane as adhesives to glue the particles together. After mixing the glue, the cork particles are molded and cold-pressed, cured by heat, and then cooled to ultimately form the cork agglomerates. The agglomerates can be then processed into sheets or blocks for direct use, or the sheets or blocks can be combined with other materials to prepare diversified cork products (Duarte and Bordado 2015, Yang et al. 2021). In addition to polyurethane glue, many studies have reported the use of epoxy resin and melamine-modified glues to prepare agglomerated cork (Antunes et al. 2020, Gil 2015, Silva et al. 2005, Song et al. 2011).

Chitosan is a copolymer that is produced by the deacetylation of chitin under alkaline conditions. It is composed of D-glucosamine and *N*-acetyl-D-glucosamine units that are connected by β-(1→4) glycosidic bonds. Chitosan has inherent antibacterial, antioxidant properties, good biocompatibility, and controllable biodegradability. The abundance of terminal amino functional groups makes it useful as an efficient adsorbent for organic- and inorganic-based pollutants. These excellent properties make chitosan is widely used in a variety of different fields, such as pharmaceuticals, wastewater treatment, and agriculture (Hamed et al. 2016, Khan et al. 2017, Philibert et al. 2017, Younes and Rinaudo 2015).

Chitosan is also soluble in acidic media and has a high charge density and strong adhesion. Several studies have explored the use of chitosan as an adhesive for wood products (Patel et al. 2013a,b, Umemura et al. 2003, Umemura and Kawai 2007). Ibrahim et al. (2013) used laccase-modified lignin to react with chitosan to prepare a non-toxic and inexpensive adhesive. Peshkova and Li (2003) reacted laccase-modified phenolic resin with chitosan to prepare adhesives with different formulations for bonding maple veneers. Patel et al. (2013c) investigated the shear strength of European pine wood bonded together by a chitosan-based adhesive, and the results showed that the best-performing adhesive consisted of 6 wt.% chitosan, 1 wt.% glycerol, and 5 $mmol \cdot L^{-1}$ trisodium citrate. Nonetheless, these reports focused on wood veneers rather than cork materials. As a part of bark, the function and main components of cork, which include suberin, lignin, polysaccharides, and extracts, are different from those of wood. Furthermore, few reports on the use of chitosan as an adhesive to prepare cork agglomerates have been reported.

The research in this paper takes advantage of the production process of wood-based panels, which utilizes hot-pressing for preparing laminated wood composites, to prepare aldehyde-free and environmentally-friendly cork agglomerates using acidified chitosan as an adhesive.

The physical and mechanical properties of the prepared cork agglomerates were tested according to standards. Then, the cork agglomerates were coated with oil-based and water-based paints to analyze and compare the film properties after painting. This research aimed to provide a technical basis for the development of the new cork agglomerate products.

MATERIALS AND METHODS

Materials

Cork particles (size range of 1- 0.5 mm, moisture content 6%) were purchased from Huacheng Cork Technology Co., Ltd (Shaanxi, China) and the moisture content was approximately 6%. Chitosan with a deacetylation degree of 80-95%, was purchased from Guoyao Group Chemical Reagent Co., Ltd (China). Glacial acetic acid (analytical grade) was purchased from Tianjin Zhiyuan Chemical Reagent Co., Ltd (China). Water-based, acrylic-modified polyurethane paint, which included a water-based sealer and water-based varnish (transparent matte), was purchased from China Nippon Paint Chengdu Co., Ltd. Oil-based polyurethane paint, which included a clear transparent primer and a clear semi-gloss varnish, was purchased from Guangdong Jiabaoli Technology Materials Co., Ltd (China).

Preparation of the chitosan adhesive

A 1 wt.% aqueous solution of acetic acid was slowly poured into a beaker containing chitosan, and the mixture was stirred at room temperature until it become completely homogeneous, forming an aqueous solution of the acidified chitosan. An insufficient amount of acetic acid caused the chitosan solution to be highly viscous, which made it difficult to spray, while excess acid diluted the chitosan solution with too much water, which prolonged the water-expelling time during the hot-pressing procedure. After optimization, it was determined that the mass ratio of the 1 wt.% acetic acid solution to chitosan was 30:1.

Preparation of the chitosan-glued agglomerated cork panels

A certain amount of the cork particles mixed with 1 wt.% chitosan glue were poured into a mold, and the matrix was cold-pressed and shaped for slab. The slab was hot-pressed at 140°C for 12 min to prepare the agglomerate, which featured a density of 0.55 $g \cdot cm^{-3}$ and dimensions of 350 × 350 × 4 mm. The thickness of the agglomerate was controlled by a pair of 4 mm thick steel bars. The hot-pressed agglomerate was cooled and cut for future tests and analysis.

Coating process

The agglomerated cork panels were polished using sandpapers with different grits (100#, 120#, 180#, 240#, and 320#) sequentially until the surface was smooth and ready for coating. The primer of the water-based, acrylic-modified polyurethane paint was directly used, but the water-based finish painting needed to be mixed with 10% distilled water. The primer of the oil-based polyurethane paint was prepared with a 1 : 0.5 : 0.4 (ratio of primer / curing agent/ thinner). The oil-based finish painting was prepared with a 1 : 0.5 : 0.5 (ratio of finish painting / curing agent / thinner). Two coats of primer (80 $g \cdot m^{-2}$ per primer coating) and two coats of finish painting (120 $g \cdot m^{-2}$ per paint coating) for water-based and oil-based paints were applied at room temperature and a relative humidity of 65 ± 3%. The tests of paint film properties were performed after the coating film completely cured. The specific coating process is diagrammed in Fig. 1.

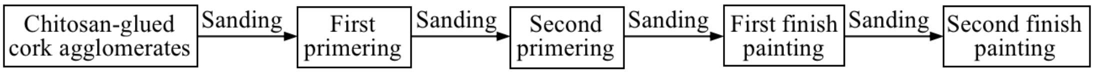

Fig. 1: Diagram of the process of coating the chitosan-glued cork agglomerates with the two types of coatings.

Physical and mechanical property tests

The boiling water resistance after cooking for 3 hours and the tensile strength of uncoated cork agglomerates were measured according to the Chinese forestry industry standard for cork flooring LY/T1657: 2015 Cork floor. Samples with sizes of 50 × 50 × 4 mm were prepared to measure the thermal conductivity and thermal diffusivity of the uncoated agglomerates using a thermal conductivity meter with a DRE transient plane heat source. The tests were repeated six times for statistical analysis.

Scanning electron microscopy (SEM)

To observe the structural characteristics of the chitosan-glued cork agglomerates, the surface of a 5 × 5 mm square piece of the uncoated cork agglomerate was flattened using a single-edged blade. Then, the flattened square was cut at the surface bonded area of cork granules to achieve a thickness of 1 mm section using a double-edged blade. The drying sections were spray-coated with gold and then were observed by field-emission scanning electron microscopy (Hitachi S-4800 SEM).

Chromaticity analysis

The color changes of the cork agglomerates before and after the two coated processes were quantitatively evaluated using a SC-80C automatic colorimeter in accordance with the CIE standard color system of the International Commission of Illumination. The experiments were conducted in six replicates per group, and the average value of the six replicate were reported. The values of L^*, a^*, and b^* were used to characterize the colorimetric properties paint, L* referred to the lightness index, a^* referred to the red-green index, and b^* referred to yellow-blue index. The values of ΔL^*, Δa^*, and Δb^* represented the difference between L^*, a^* and b^* before and after painting, and ΔE^* represented the total color difference, which was used to quantify the difference in color perception. Smaller ΔE^* values represented smaller color changes.

Glossiness of the cork agglomerates

A 60° Glossmeter was utilized to evaluate the glossiness of cork agglomerates before and after coating, using the China standard GB/T4893: 2013 Test of surface coatings of furniture, as a reference. Since the surface of the cork agglomerates did not have a texture direction, the tests were performed along the center parallel to the random edge of the sample.

The glossiness measurements were repeated 6 times for each group, after which the average value was calculated.

Adhesion classification

A BN709 multifunctional paint film detector was employed to measure the paint film adhesion of cork agglomerates coated by water-based, acrylic-modified polyurethane paint and oil-based polyurethane paint, using the China standard GB/T4893: 2013 as a reference. The cross lines of paint film on the coated cork agglomerates were cut using cutter knife, and the cross lines were observed and evaluated with a magnifying glass.

Abrasion resistance

The abrasion resistance of the paint film of coated cork agglomerates was determined by a Taber abrasion resistance tester, using the China standard GB/T4893: 2013 as a reference. The paint films after grinding 100 revolutions at the turntable speed of 60 rpm were evaluated and the mass loss (F) was expressed by Eq. 1:

$$F = \frac{m - m_1}{m} \times 100\% \quad (1)$$

where: F - mass loss of sample after grinding 100 revolutions (%), m - the mass of sample before grinding (g), m_1 - the mass of sample after grinding 100 revolutions.

RESULTS AND DISCUSSION

Physical and mechanical properties of uncoated cork agglomerates

The chitosan-glued cork agglomerates prepared, which featured a density of 0.55 $g \cdot cm^{-3}$ and a thickness of 4 mm, boasted an average tensile strength of 1.70 MPa when the cork particles were glued together using 1.0 wt.% chitosan by hot-pressing at 140°C for 12 min. The measured tensile strength was greater than 1.4 MPa, meeting the requirements of the LY/T1657: 2015 standard concerning the tensile strength of cork-based materials. The agglomerated cork panels were not loosened even after immersion in boiling water for 3 hours, which was also a requirement of cork-based materials based on the LY/T1657: 2015 standard. The thermal conductivity of the cork agglomerate was 0.11 $W \cdot m^{-1} \cdot K^{-1}$, and the thermal diffusion coefficient was 0.13 $mm^2 \cdot s^{-1}$.

Wei et al. (2019) used polyurethane adhesive as the binder to prepare the cork floor. The optimal process parameters of cork floor were 9 wt.% resin content, 120°C hot pressing temperature and 12 min hot pressing time. The maximum tensile strength of cork floor was 1.60 MPa. Antunes et al. (2020) showed that an average tensile strength of 1.50 MPa when cork-based panels with a density of 0.60 $g \cdot cm^{-3}$ and a thickness of 2 mm were glued together in a deformable aluminum mold using 3.8 wt.% melamine-urea-formaldehyde (MUF) by hot-pressing at 150°C for 5 min and a cooling time sufficient to achieve 25°C. The tensile strength of cork-based panels increased with resin content. MUF resin yielded a considerably higher tensile strength than polyurethane resins.

Compared with the above research results, chitosan-glued could be used to prepare agglomerated cork panels under the optimized hot pressing parameters in this research. Paiva et al. (2016) also used acidified chitosan as natural adhesives for cork and the glued cork discs yielded the best tensile strength. Chitosan was able to establish electrostatic interactions, hydrogen bonding, and van der Waals forces between D-glucosamine and the adherend. When chitosan was dissolved in an acidic solution, the amino functional groups in the chitosan molecule became protonated, providing the macromolecule with an abundance of positive charges that facilitated strong covalent interactions with -COOH within the cork matrix (Peter 1995).

SEM

The SEM images of the chitosan-glued cork agglomerates are shown in Fig. 2. The cork particles that comprised the cork agglomerates spatially were organized tightly (Fig. 2a). In addition, most of the cell lumens of cork particle surface were obviously compressed, deformed, or even squeezed together, and the cork cell walls were more obviously wrinkled after compression (Fig. 2b). Under the setting density (0.55 g·cm^{-3}) and laboratory conditions, the different cork particles had different degrees of compression, and the surface featured micron-sized gaps between cork particles as well as some uncompacted cell lumens. These enabled the water-based or oil-based paint to form glue nails on the cork agglomerates after being coated.

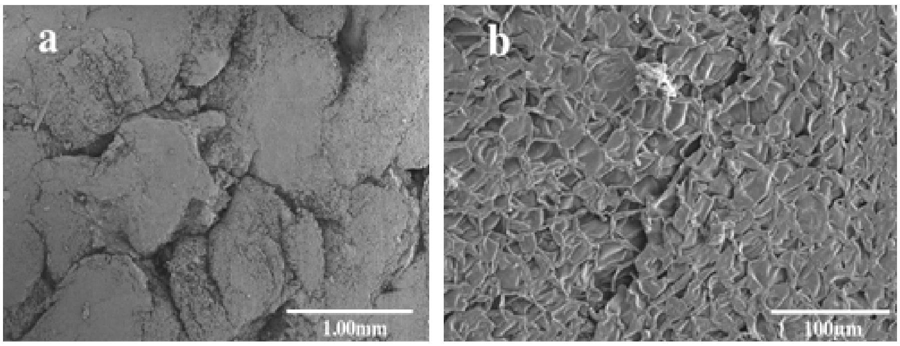

Fig. 2: SEM images of the chitosan-glued cork agglomerates.

Color differences after painting

The changes of chromaticity parameters of the cork agglomerate samples coated with the two different types of paints are shown in Tab. 1. The photos of products are shown in Fig. 3.

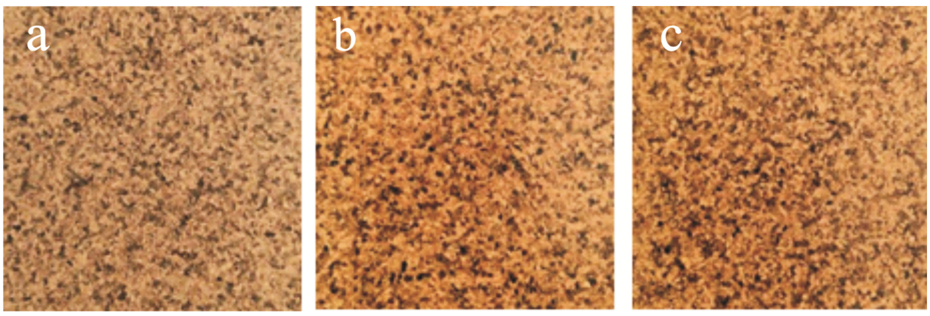

Fig. 3: Images of the chitosan-glued cork agglomerates before and after coating: (a) control, (b) coated with the oil-based polyurethane paint, (c) coated with water-based, acrylic-modified polyurethane paint.

Tab. 1: Chromaticity parameters of the chitosan-glued cork agglomerates before and after coating.

Painting type		L^*	a^*	b^*	ΔL^*	Δa^*	Δb^*
Oil-based polyurethane	before	51.71	12.19	23.06	—	—	—
	after	40.38	13.21	24.26	-11.33	1.02	1.20
Water-based, acrylic-modified polyurethane	before	51.73	11.04	22.35	—	—	—
	after	39.71	12.15	23.17	-12.02	1.11	0.82

The chromatic parameter variation is related to the paint type and coating process (Hang et al. 2020). The $L*$ value of the cork agglomerate decreased from 51.71 to 40.38 after being coated with the oil-based polyurethane paint, while the $a*$ value increased from 12.19 to 13.21, and the b* value increased from 23.06 to 24.26. On the other hand, after coating the agglomerates with the water-based, acrylic-modified polyurethane paint, the $L*$ value of the cork agglomerate decreased from 51.73 to 39.71, the $a*$ value increased from 11.04 to 12.15, and the $b*$ value increased from 22.35 to 23.17. These results illustrated that the surface brightness of the cork substrate decreased after being coated, while the red-green and yellow-blue indexes both slightly increased.

The total color difference ΔE^* is a numerical way to express the difference in color perception of two colors. The change in ΔE^* of cork agglomerates before and after coating had a certain relationship with the composition of the paint and the coating process. According to the color difference formula ($\Delta E^* = (\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2})^{1/2}$), the calculated ΔE^* of oil-based polyurethane paint and the ΔE^* of the water-based, acrylic-modified polyurethane paint were 11.44 and 12.09, respectively. The ΔE^* of cork agglomerates before and after coating both had marked change because of the large change in lightness.

Gloss of paint films

The glossiness of a coating film reflects the reflectance of light off the surface of the film. For a biomass substrate, a higher degree of gloss leads to a better visual effect. Tab. 2 shows the recorded gloss values after shining light at a 60° angle of agglomerated corks before and after coating. The gloss value of the uncoated cork agglomerate increased substantially from 1.97 to 36.72 at 60° after being coated with the oil-based polyurethane paint. Another uncoated cork agglomerate displays a gloss value of 1.90 at 60°. After being coated with the water-based, acrylic-modified polyurethane paint, the gloss of the cork agglomerate reached 27.99. These changes indicated that both the oil-based polyurethane paint and the water-based, acrylic-modified polyurethane paint endowed the cork agglomerates with higher gloss compared to the uncoated cork agglomerate. Because the agglomerated cork comprised small particles, most of the cork cells on the surface of the particles exposed the cell lumens with varying degrees of compression after hot pressing. The uncoated surface of agglomerated cork featured some grooves and uncompacted cell lumens that readily diffused the reflected light, resulting in lower gloss. The paint coatings easily filled in some uncompacted cell lumens, forming a flat film on the surface after curing. Therefore, two kinds of paint coating endowed the cork agglomerate with significantly higher gloss. The variety of paints has a large impact on gloss, which is due to the difference of pigment particle size, pigment-base ratio and dispersion in the base material of different coatings. In the same coating process, the glossiness of all samples coated with the oil-based polyurethane paint was higher than those coated with the water-based, acrylic-modified polyurethane paint.

Tab. 2: Gloss values of the chitosan-glued cork agglomerates with and without coatings measured at 60°.

Sample		1	2	3	4	5	6	Avg.
Oil-based polyurethane paint	before	1.47	2.40	2.12	1.97	2.30	1.57	1.97
	after	36.57	34.62	38.68	37.87	38.55	34.02	36.72
Water-based, acrylic-modified polyurethane paint	before	1.56	2.27	1.97	1.67	2.25	1.66	1.90
	after	26.75	28.45	28.03	28.78	26.87	29.07	27.99

Adhesion of paint films

Adhesion is an important index of paint film and plays a prominent role in guiding the coating process of agglomerated cork panels. Fig. 4 displays images of the adhesion test of the coated cork agglomerates. As shown in Fig. 4 and Tab. 3, a smooth cutting edge without shedding and strong adhesion resulted after cutting the cork agglomerate coated with the oil-based polyurethane paint; therefore, the oil-based polyurethane paint was given a 0 rating: the highest adhesion level. The cork agglomerate coated with the water-based, acrylic-modified polyurethane paint featured a small amount of coating that peeled off at the intersection of the incision. However, the affected cross-section was no more than 5% of the total area; this adhesion of paint film was rated as a level 1.

The SEM images in Fig. 2b demonstrated that there were many exposed cell lumens at the surface of the cork agglomerates. After the agglomerates were coated with the two paints, the liquid coatings filled in the gaps between the cork particles and cell lumens which were not completely compressed, forming glue nails after curing that promoted mechanical interlocking and, therefore, strong adhesion of the coating to the cork agglomerate. Based on the results of the adhesion tests, the adhesion of the oil-based polyurethane paint to the cork agglomerate was stronger than the water-based, acrylic-modified polyurethane paint.

Tab. 3: Evaluation of the adhesion level of the two different cork agglomerates coatings.

Sample	1	2	3	4	5	6	Level
Oil-based polyurethane paint	0	0	0	0	0	0	0
Water-based, acrylic-modified polyurethane paint	1	1	1	1	1	1	1

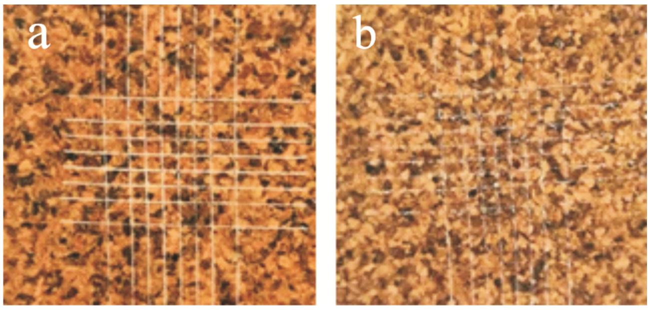

Fig. 4: Images of the chitosan-glued cork agglomerates during the adhesion tests: (a) oil-based polyurethane paint coating and (b) water-based, acrylic-modified polyurethane paint coating.

Abrasion resistance of paint films

In addition to nature of paint itself, the abrasion resistance of paint films is also related to the amount of finish coating. In accordance with the referenced paint film abrasion resistance standard, cork agglomerates coated with the oil-based polyurethane paint and water-based, acrylic-modified polyurethane paint were polished for 100 revolutions at the turntable speed of 60 rpm. Images of the cork agglomerate samples being polished for 100 revolutions at the turntable speed of 60 rpm by the abrasion tester are shown in Figs. 5 and 6.

After undergoing abrasion for 100 revolutions, the cork agglomerate coated with the oil-based polyurethane paint exhibited a mass loss of 1.39%. The cork agglomerate substrate did not expose after undergoing abrasion for 100 revolutions. Comparably, the cork agglomerate coated with the water-based, acrylic-modified polyurethane paint underwent a mass loss of 0.81% after enduring abrasion for 100 revolutions. The cork agglomerate substrate also did not expose after grinding 100 revolutions.

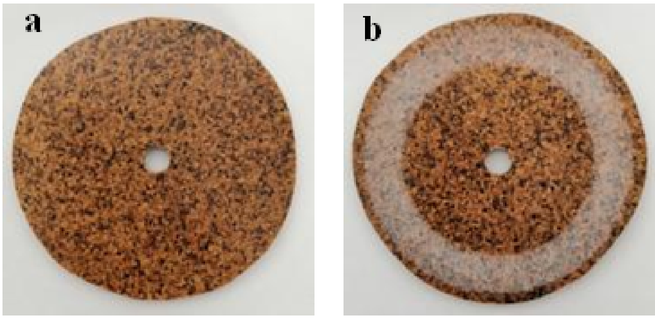

Fig. 5: Undergoing abrasion on the integrity of the oil-based polyurethane paint:(a) before abrasion, (b) polished for 100 revolutions at the turntable speed of 60 rpm.

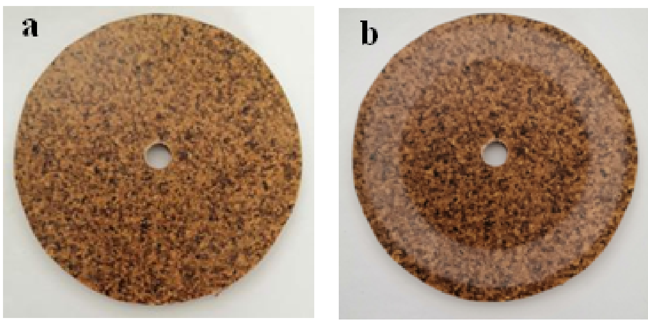

Fig. 6: Undergoing abrasion on the integrity of the water-based, acrylic-modified polyurethane paint: (a) before abrasion, (b) polished for 100 revolutions at the turntable speed of 60 rpm.

In this research, chitosan was used as the main component to synthesize environmental friendly adhesive, which had been successfully applied in preparation of agglomerated cork panels by hot-pressing. The properties of acidified chitosan adhesive, such as viscosity, active period and storage period have great influence on the performance of cork agglomerates, and need further exploration. The property of the paint, coating process and cork substrate have an impact on the properties of paint film. The surface decoration and visual effect of agglomerated cork panels had been improved after coating oil-based polyurethane paint and water-based, acrylic-modified polyurethane paint. The choice of paint needs to be considered according to the use of cork panels. The acrylic-modified polyurethane paint with water as solvent has the advantages of simple formulation, non-toxic and greenness, and has great market potential and application prospect in cork industry. However, the contents about the influence of ambient temperature and humidity changes on the paint film properties, the drying cost and aging resistance of paint film and so on, still need further research.

CONCLUSIONS

(1) The acidified chitosan demonstrated to be a successful adhesive for agglomerating boards made of cork particles. The optimal mass ratio of acetic acid (1 wt.% in distilled water) to chitosan was found to be 30:1. *(2)* The resulting cork agglomerates featured a density of 0.55 $g \cdot cm^{-3}$ and a thickness of 4 mm, and they demonstrated a tensile strength of 1.70MPa, a thermal conductivity of 0.11 ($W \cdot m^{-1} \cdot K^{-1}$), and a thermal diffusion coefficient of 0.13 $mm^2 \cdot s^{-1}$. The agglomerated cork panels were not loosened after immersion in boiling water for 3 hours. *(3)* The cork agglomerates coated with either the oil-based polyurethane paint or the water-based, acrylic-modified polyurethane paint displayed decreased lightness and increased gloss compared to the uncoated agglomerates. In terms of adhesion of paint films, the oil-based polyurethane coating was rated an adhesion level of 0 (highest), while the water-based, acrylic-modified polyurethane coating was rated as a level-1 adhesion. The substrates of cork agglomerates coated two types of paint did not expose after undergoing abrasion for 100 revolutions at the turntable speed of 60 rpm.

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