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Fungicide and insecticide properties of cardboard panels made from used beverage carton with veneer overlay

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This study evaluated fungicide and insecticide properties of the cardboard substrate panels overlaid with beech veneer. The experimental cardboards from recycled food and beverage carton containers having approximately 75% paperboard, 20% low density polyethylene (LDPE), and 5% aluminum foil were overlaid using four types of adhesives; polyurethane (PU), phenol–formaldehyde(PF), urea–formaldehyde(UF) and melamine–urea formaldehyde (MUF). The cardboard specimens overlaid with veneer using polyurethane adhesive had better mechanical properties and water resistance than those of the specimens made with other three types of adhesives. Brown rot fungus *Coniophora puteana* and white rot fungus *Ceriporiopsis subvermisphora* were used for the decay tests. The larvae of the *Rhagium bifasciatum* F. was used for the insect test. Wood veneer faced cardboards had significantly higher antifungal and insecticide properties than those of the control wood samples. The weight losses of the cardboard groups caused by *C. puteana* and *C. subvermisphora* were 0.43 - 0.83%. While UF-cardboard type board was the most affected group by the fungus, MUF-cardboard type was found as the most resistance group against larvae.

Key words: Antifungal, insecticide, composite, recycling, used beverage carton, wood veneer.

INTRODUCTION

In recent years, environmental problems and recycling issues are being discussed with more popularity in most of the developed and developing countries. In 2006, 313000 tons of beverage carton were recycled within a total capacity of 12 billion tons recycled material that represents a recycling rate of 30% in Europe. Recycling is not only increasing at a high rate but also combining with recovery of material reaching to almost 636000 tons with an approximate value of 61% rate in European Union. It is expected that more than 70% of municipalities will have enhanced opportunities for recycling household packaging (Anonymous, 2007a). Recycled food carton also has substantial amount of market share within recycling industry.

In recent years, environmental problems and recycling issues are being discussed with more popularity in Turkey. Recovering waste material from used beverage cartons (UBC) to manufacture a value-added product with an economical and efficient method is an important issue from the perspective of environmental pollution. Recycling of beverage cartons is a relatively new developing industry in Turkey. The material obtained from recycled UBC carton in Europe is predominantly used for the manufacture of paper and carton based products including shopping bags, cores for paper reel, sheets of cardboard, disposable kitchen towels, printing paper, plaster board lining and corrugated board (Anonymous, 2007b). Throughout the manufacturing process, the cartons are treated in such a way that no other material including toxic adhesives are needed. Cardboard also offers excellent soundproofing and insulation qualities

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- 1. Polyethylene: barrier to moisture, bacteria
- Paperboard: for stiffness
- Polyethylene: adhesion layer
- Aluminum: barrier to oxygen, light
- Polyethylene: adhesion layer
- Polyethylene: seals in liquid food contents

Figure 1. The dominating structure of aseptic beverage cartons (Buelens at al, 2001).

(Ayrilmis et al., 2008).

UBC carton can be recycled using a thermal compression process to manufacture home and building products. The dominating structure of a used beverage carton is seen in Figure 1. As an alternative to repulping for paper applications, an additional process converts shredded cartons into thermally compressed to make a high strength bio-composite panel alternative to traditional wood based panels such as particleboard, medium density fiberboard (MDF) and oriented strandboard (OSB). This type of panel product was developed by Tetra Pak[®] and is produced in various countries under the brand name (Ayrilmis et al., 2008).

Yekpan[®] is composed of 70 - 90% paper, 10 - 25% low density polyethylene (LDPE) and about 5% aluminum which are existing components of UBC produced in Turkey. UBC cartons collected from consumers are shredded and then molded together under high temperature and pressure. The process uses the whole waste package without leaving any waste. UBC cartons are shredded into 5 mm particles and formed in a layer to get a desired thickness. The mat is then compressed under pressure and heat in a hot press (Buelens et al., 2001). In this process, there is no need for the addition of an adhesive element due to 20% polyethylene in UBC carton raw material. The polyethylene content in the mat melts and binds fiber and aluminum pieces together in the form of a compact elastic matrix. Aluminum at the rate of 5% causes the heat to spread more uniformly (Ayrilmis et al., 2008).

Overlaying of wood based panels such as particleboard and MDF with wood veneer sheets improves their appearance and properties resulting in value-added products. Veneer overlay is a Premium product in cabinet and furniture manufacture, as far as its quality is concerned. Although particleboard provides an excellent surface for the application of quality wood veneer, welldeveloped adhesive strength between overlay and substrate is required to have an ideal lamination process (Hiziroglu and Rabiej, 2005). Currently, interior fitness and furniture manufacturers using wood based panels such as particleboard and MDF overlaid with decorative surfacing material, do not commonly know cardboards made from UBC chips. However, when cardboards are overlaid with decorative wood veneer sheets using a suitable adhesive, they could be a competitor to overlaid wood based panels in office furniture manufacture. These overlaid panels could be used in the construction of cabinets, furniture, paneling, kitchen worktops and work surfaces in offices, educational establishments, laboratories and other industrial product applications (Ayrilmis et al., 2008).

Although many types of wood are relatively easily deteriorated by a variety of organisms, including fungi and insects, in recent years composite wood materials have received more attention than solid wood because of changes in building design and protective practice. While they show a greater resistance to biodeterioration, they are still susceptible to biological attack. In most cases, these wood-based materials perform well as long as they are used in dry conditions, but they are increasingly used where they are likely to become wet and will ultimately deteriorate. A number of protective methods allow these materials to perform more reliably under conditions suitable for biodeterioration (Kartal and Ayrilmis, 2005).

Physical and mechanical properties of the cardboard substrate panels overlaid with beech veneer have recently been evaluated as part of a collaborative project between the Forestry Faculty of Istanbul University, Turkey and Forestry Faculty of Oklahoma State University (Ayrilmis et al., 2008). In this study reported, another part of the project, investigated fungicide and insecticide resistance (decay resistance) of the cardboard panels overlaid with beech veneer. Determining of the adhesive type having the best decay resistance among experimental adhesives was also aimed.

MATERIALS AND METHODS

Preparation of test specimens

Four commercially produced cardboard panels with dimensions of $1250 \times 2500 \times 15$ mm were supplied by Yekas Recycling Company. The panels were then cut into smaller test panels with



b) Top



Figure 2. A cardboard sample overlaid with beech veneer sheets.

dimensions of 500 × 500 × 15 mm. A total of 20 experimental panels were randomly assigned to experimental groups, four for each type of adhesive and four panels without overlay application. Commercially produced defect free rotary-cut beech (Fagus orientalis L.) veneer sheets with a thickness of 1.5 mm and an average density of 0.63 g/cm³ were used to overlay cardboards (Berkel, 1970). All of the experimental panel parts and veneer sheets were placed in a climate room with a temperature of 20 ± 2°C and a relative humidity of 65 ± 5% before overlaying process was carried out.

Cardboard panels were manufactured at Yekas Recycling Company, a recycling institution acquired to Turkey by Tetra Pak[®], Izmir, Turkey. Raw material was shredded into 5 mm particles before the paperboard-LDPE (low density polyethylene)-aluminum particles were then spread into sheets to a desired thickness. Mats without having any resin or additives were pressed in a hot press at a temperature of 170 °C and a pressure of 1.2 N/mm² for 12 min as illustrated in Figure 2. No adhesive was used since there is 20% polyethylene which melts and acts as an adhesive to bond the board in UBC. Aluminum at the rate of 5% causes the heat to spread more uniformly during the press. The panels were then cooled in cold platforms.

Overlaying process of the cardboard with beech

Two pieces of 1.5 mm thick veneer sheets were glued on both sides of the cardboard (Figure 2). The face veneers were aligned to the cardboard substrate so that their longitudinal grain direction was perpendicular to the major axis of the substrate cardboard. Four types of liquid synthetic adhesives were used to bond the veneer sheets to the cardboards, urea-formaldehyde (UF), phenolformaldehyde (PF) and melamine/urea-formaldehyde (MUF) with a mixture of 20% melamine, 80% urea and polyurethane (PU) (diphenylmethane-4,4'-di-isocyanate). Ammonium chloride (NH₄Cl) was added as a hardener in the case of UF and MUF adhesives at a level of 1% based on dry resin weight. Each adhesive were uniformly applied on each side of the face veneers at approximate rate of 200 g/m². The panels were then sandwiched with the veneer

sheets and pressed at a pressure of 1.5 N/mm² and a temperature of 120 °C for 10 min in a laboratory type hot press except for panels made using PU resin. The press parameters of PU adhesive were 1.5 N/mm² and 60 °C for 180 min for pressure, temperature and time, respectively. Finally, the experimental cardboards with veneers were conditioned in a climate chamber with a relative humidity of 65 ± 5% and a temperature of 20 ± 2°C for about 3 weeks before being cut into test specimens.

Fungus tests

The veneer faced cardboard and control (solid wood) samples for the fungus tests were placed in petri dishes containing agar medium inoculated with white rot fungi, Ceriporiopsis subvermisphora, brown rot fungi and Coniophora puteana according to standard (TS 5563 EN 113 -1996). All samples were incubated for 12 weeks (25°C and 65 ± 5 for C. puteana; 27°C and 90% humidity for C. subvermisphora).

Insect test

Test samples were exposed to insect larvae (two, weekly) of Rhagium bifasciatum Fabr. (Longhorn Beetle). Six samples with dimensions of 50 x 25 x 15 mm were prepared according to standard (TS 5564 EN 47-1996). Besides, eight control samples for comparing to the cardboard samples were prepared from Scots pine sapwood. Test samples were drilled 10 mm diameter lengthwise from central by using drill. The larvae were placed into the samples, then holes were closed by cotton. For respiration of larvae, a piece of cotton was used while closing the sample holes. The living of larvae was observed for 12 weekly test period. Insect tests were made according to the standard method (TSE 5566 EN 22). The incubation conditions were the temperature of $27 \pm 2^{\circ}$ C and humidity of 80 \pm 5%. Air circulation was also applied for two hours every day in the room. Tests samples were examined after a 3-months fungal decay and insect tests.



Figure 3. The weight losses of cardboard samples in terms of test fungus (a) and chemicals used (b).

Source	Sum of squares	Df	Mean square	F	Sign
Fungus species	0.309	1	0.309	3.151	0.083
UBC board groups	0.174	3	0.058	0.591	0.625
Fungus spec * Fruit boxes board groups	0.360	3	0.120	1.226	0.313
Error	3.920	40	0.098	-	-
Corrected Total	4.763	47	-	-	-

Table 1. ANOVA table for weight losses of test samples after fungal decay.

Dependent variable: Based on oven dry weight. $R^2 = 0.177$ (adjusted $R^2 = 0.033$).

Fungus and insect test results were evaluated using a computerized SPSS 13.0 statistical program and tested with univarite analysis, followed by a Duncan test with a 95% confidence level.

RESULTS AND DISCUSSION

Decay test results

The weight losses of the cardboard and control samples in terms of adhesive types between wood veneer and cardboard surface was shown in Figure 3. The weight losses of the board samples were in minimum level, compared with Scots pine control samples. The weight losses of the MUF-, PU-, PF- and UF-cardboards exposed to *C. puteana* fungi were found as 0.43, 0.47, 0.69 and 0.47% respectively, while they were found for *C. subvermisphora* fungi as 0.72%, 0.83%, 0.62%, and 0.54%, respectively. The weight losses of Scots pine control samples was 18.6% for *C. puteana* inoculated samples and 19.3% *C. subvermisphora* inoculated samples.

There were no significant differences among weight losses of the cardboard samples (Table 1). However, the weight losses and larvae living rate of the Scots pine samples were significantly higher than those of the cardboard samples. The one way and two way interactions of fungus species and the cardboard groups were not significant at the 95% confidence level.

Significant differences between mean values of the cardboard groups were determined using Duncan's multiple range test (Table 2). The cardboards with similar antifungal activity were indicated by the same letter in homogeneity cardboard groups (Figure 3). The veneer faced cardboards made using UF, M, PU and PF to show the same antifungal activity with rate of weight losses

Cardboard groups	Weight losses with Cp	HG	Cardboard groups	Weight losses with Cs	HG
MUF	0.4300	a*	UF	0.5058	s*
PU	0.4683	а	PF	0.5758	S
UF	0.4733	а	Μ	0.6475	S
PF	0.6900	b	PU	0.6533	S
Significant	0.092		Sig	0.237	
Control	18.6%			19.3%	

Table 2. Duncan test results of the groups concerned with average weight losses of particle board groups.

Cp; Coniophora puteana, Cs; Ceriporiopsis subvermisphora, HG; Homogeneous groups.

* Means within each factor and column followed same letter are not significantly different (α =0.05).

 Table 3. The last states of the larvae and mature insect at the end of the test.

Condhoord groups	Total larvae	Living larvae		Mature insect		Mortality rate	
Caroboard groups		Number	%	Number	%	Number	%
MUF	8	0	0			8	100
PF	8	1	12.5	5	62.5	2	25
PU	8	4	50			4	50
UF	8	2	25	5	62.5	1	12.5
Scots pine control	8			8	100	0	0

0.5 - 0.65% for both of fungus species.

Insect test results

The last states of the larvae and mature insect at the end of the test was shown in Table 3. The results are concerned with insecticide features in the test samples observed and are very different from the results of fungicide features.

Some differences between board groups and sub groups same boards were determined. The larvae could not gnaw hard the cardboards samples. However, some larvae could live for a long time without any gnaw-dust and did not transit to pupa period in the humid test samples which had high toxic effects. Most of the larvae in the cardboard samples had attempted to escape out of the test samples permanently. It is observed that, the larvae were not adopting any cardboard sample as nutriment sources. A typical larvae is shown in Figure 4.

Transition to pupa period of the larvae in the test samples had started when the age of larvae was 4 - 6 week. All of the larvae mortality in the samples began at the first week. The larvae's transition to pupa period had no need to be nourished from the samples. Thus, the larvae had mature individuals without influence by toxicities of samples. The larva and pupa period from the 6th week to 12th week, was accepted as a mature individual according to TS 5563 standard. Significant differences relating to toxic effects between cardboard groups were determined individually by Duncan's multiple-comparison tests as shown in Table 4.

The toxicity of sample was not enough for the larvae attending their live as chrysalis period in the cardboard. Toxic effects of the boards against larvae were shown in Figure 5.

The observations concern with larvae

Any larvae did not gnaw the cardboard samples due to the hardness of the samples. Any gnaw-dust in the cardboard samples was not observed. Most of the larvae in the cardboard samples had attempted to escape out of the samples. They had turned towards cotton which used plug permanently for being nourished. Fifteen percent of the larvae died and rest of them are attending their lives as larvae in cardboards made with PU resin, while the cardboards made with MUF resin had a high toxicity for the larvae, all larvae died in the samples in the first week. 60% of the larvae could be mature individuals in the samples made with PF and UF resins. One larva in the samples made with PF resin and two in the samples made with UF resin were attending their live as larvae. This larva was alive at the end of 12 weeks, but any gnaw-dust in the test sample was not determined. Other larvae had died in the first two weeks. All larvae formed gnaw-dust for nourishment and pupa term which was enough amounts in the Scots pine control samples. The amount of dust of pupa pillow and the period of the pupa to mature were similar.



Figure 4. Rhagium bifasciatum larvae in MUF-cardboard sample.

Table 4. The classification in the cardboard groups in view of larvae living rate obtained from Duncan test.

Cardboard groups	Larvae living rate %	HG
MUF	0.2708	а
PU	0.6354	b
PF	1.5208	С
UF	1.6563	С
Scots pine control	2.0104	d
	Cardboard groups MUF PU PF UF Scots pine control	Cardboard groups Larvae living rate % MUF 0.2708 PU 0.6354 PF 1.5208 UF 1.6563 Scots pine control 2.0104

HG: Homogeneous groups, α =0.05



Figure 5. Toxic effects of the cardboard panel samples against larvae (%).

Conclusion

The results of the present study showed that raw material from recycled cardboard can be used for manufacture of composite boards with veneers sheet. Both antifungal and insecticide properties of the samples resulted in satisfactory values meeting minimum requirements stated in TS 5564 EN 47 1996, TS 5563 EN 113/1996, TS 5565 EN 22 1996. Wood veneer faced cardboards had significantly higher antifungal and insecticide properties

than those of the control wood samples.

The cardboards made from used beverage carton had a high resistance to fungus and test insect R. bifasciatum. While cardboard overlaid veneer made using UF resin was the highest affected group by the fungus and insect, cardboard overlaid made using MUF resin was found as the highest decay resistance and the toxic effect among all cardboard samples against larvae. For this reason, MUF-cardboard group can be used for heavy exposure conditions as an alternative to solid wood. Test results informed us that MUF-cardboard group was not a good nourishment sources for the larvae. Because cardboards had a toxic effect to larvae, larvae intended to escape from the all cardboard groups. Test results revealed that cardboard can be used as a substrate with wood veneer sheets in heavy humidity conditions such as bathroom furniture and roofing material which are exposed to fungus and insect risk areas. In addition, panels made from recycled cardboard have lower production cost than those of wood based panels since raw material for cardboard panel manufacture is widely available and has low cost as compared to wood raw material.

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