

Composite boards produced from waste Tetra Pak® packaging materials: Chemical properties and biological performance

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Abstract

Developing new materials with improved performance by using various biomass sources and wastes has been received much attention in recent years due to several benefits such as environmental and socioeconomics. Composite panels from waste Tetra Pak® cartons, a worldwide well-known multi layer beverage packaging system composed of cellulose, low density polyethylene (LDPE) and aluminum, were manufactured. In the study, fungal decay and termite resistance tests were performed in laboratory conditions along with tests for mold resistance to evaluate biological performance of the panels. Thermal degradation properties were determined by thermo-gravimetric (TG) and differential thermo-gravimetric (DTG) analyses. Semi-quantitative elemental analyses were run by energy-dispersive x-ray analysis along with scanning electron microscope (SEM). Besides recycling in pulping, this study showed that it is possible to use waste Tetra Pak® cartons to manufacture panels without leaving any waste. Fungal decay resistance tests revealed that the panels were not resistant wood-decaying fungi; however, the panels were resistant against mold fungi and termites. Thermal degradation reactions of paper layer itself occurred at lower temperatures than those occurred in both LDPE layer itself and the panel. While thermal degradation reactions of paper layer began to occur at temperatures between 200-350°C, thermal degradation of LDPE layer started at 432°C. Since aluminum does not degrade during thermal treatment and melts at around 600°C, it seems that it has no effect on the trend of TG and DTG profiles of the panels. Such panels may have limited usage areas regarding to hydrophilic character of cellulose in the structure and indoor applications where fungal decay risks are absent can be considered for the panels without any additives.

Keywords: fire, fungus, panel, termite, Tetra Pak, waste cartons.

Introduction

Tetra Pak® is the best-known and largest producer of beverage cartons with an annual worldwide production of about 150 billion packages. Due to huge amount of consumption of such products, considerable amount of the cartons become waste material every year. Recycling of postconsumer packaging is of both environmental and economical concerns. Tetra Pak® packaging cartons are made of three different materials: duplex paper (75% -cellulose fibers), low-density polyethylene (20%) and aluminum (5%). Paper material used in Tetra Pak® packaging is unbleached sulfate (kraft) and CTMP-(chemi-thermo-mechanical process) pulp.

Due to possibility of food-contact, no recycled fibers are used in manufacturing Tetra Pak[®] packages. Even 25% of the packages are residuals for paper mills, re-pulping still stands the most common method for recycling of Tetra Pak[®] packages. Cellulose fibers are separated from the polyethylene and aluminum layers in this process and fibers are used to produce cardboards, corrugated paper and new paper products. Besides re-pulping, manufacturing panels is another way to utilize waste Tetra Pak[®] packaging cartons. The advantages of making panels over pulping and separating layers are less energy demand, low cost, less complicated process for recovery and usage of whole material without leaving any waste. During the recovery process, recycled fibers wear out and are inevitably lost; however, producing panels could be useful for a long time.

Previous studies suggested that panel products from waste cartons could be an alternative raw material in the furniture industry by using suitable adhesive due to its improved mechanical properties, increased water resistance, and high biological performance against decay fungi and insects (Ayrilmis et al., 2008; Sen et al., 2010). An attempt has been made here to evaluate biological performance and thermal degradation of panels produced from waste Tetra Pak[®] cartons without any additives to present an overview regarding possible usage areas of such panels.

Material and Method

Manufacturing panels from waste Tetra Pak[®] cartons

Shredded waste Tetra Pak[®] packaging cartons were supplied from Yekasan Yeniden Kazanma A.S. converting factory in Izmir, Turkey. A blend of shredded particles without any biocide or additives was compounded in a closed mixing blender with a constant blender revolution of 30 rpm. Panels were made using a mold (60 mm by 60 mm) that was hot pressed at 170-180°C for 5-7 min at 25 kg/cm² and then cooled at room temperature. The resulting panels were cut to 50 mm by 50 mm by 18 mm.

Decay resistance tests – AWWA E10-09

Panel specimens (19 mm by 19 mm by panel thickness) were tested against two brown rot and two white rot fungi in soil block tests based on the AWWA E10-09 standard method (AWWA, 2010). In the tests, southern pine feeders were inoculated with the brown rot fungi *Postia placenta* (Fries) Lars. & Lomb. MAD 698, and *Gloeophyllum trabeum* (Pers:Fries) Murrill, MAD 617, and maple feeders were inoculated with the white rot fungus, *Irpex lacteus* HHB 7328 and *Trametes (Coriolus) versicolor* (L.: Fr.) Quel. MAD 697. The cultures were obtained from USDA Forest Service, Forest Products Laboratory, Madison, WI, USA. All cultures were maintained on malt-agar medium in Petri dishes at 27±2°C until needed. A piece of fungus inoculum equivalent to approximately 10 mm square from near the leading edge of mycelium in Petri dish cultures was cut. Sections of inoculum were placed on an edge of the feeder strip on the soil. The culture bottles with lids released one-fourth turn from a tightened position were incubated at 27°C and 70% RH for 3 weeks until the fungus completely colonized each feeder. Following conditioning period, specimens were steam-sterilized and placed on actively growing feeders in soil bottle cultures. Soil bottles were incubated at 27°C, 70% RH for 12 weeks. Following incubation, surface mycelium was brushed from each specimen before the specimens were oven-dried at 40°C for 24h, and reconditioned at 27°C and 70% RH to a constant weight. Average percentage mass loss was calculated for specimens in each treatment group.

Termite resistance – JIS K 1571

Panel specimens (20 mm by 20 mm by panel thickness) were exposed to the subterranean termites, *Coptotermes formosanus* Shiraki, according to the JIS K 1571 standard method (JIS, 2004). An acrylic cylinder (80 mm diameter, 60 mm height) whose lower end was sealed with a 5 mm thick hard plaster (GC New Plastone, Dental Stone, GC Dental Industrial Corp., Tokyo, Japan) was used as a container. A test specimen was placed at the centre of the plaster bottom of the test container. A total of 150 worker termites collected from a laboratory colony of Research Institute for Sustainable Humanosphere (RISH), Kyoto University, Japan were introduced into each test container together with 15 termite soldiers. The assembled containers were set on damp cotton pads to supply water to the specimens and kept at 28°C and >85% RH in darkness for three weeks. The mass losses of the specimens due to termite attack were calculated based on the differences in the initial and final oven-dry (60°C, 3 days) weights of the specimens after cleaning off the debris from the termite attack.

Termite resistance tests – AWWA E1-06

A no-choice termite resistance test with *Reticulitermes flavipes* Kollar (Eastern subterranean termites) was performed using five test specimens (20 mm by 20 mm by panel thickness). One specimen was placed in a glass container with moist sand and one gram of *R. flavipes*. The containers were maintained at 25°C and 80% RH for 4 weeks based on a modified AWWA E1-06 standard method (AWWA 2010). At the end of the test, specimens were oven dried and reweighed to determine mass loss. Termite mortality occurring at the end of the 4-week-exposure period was also recorded.

Mold resistance tests – ASTM D4445-10

Panel specimens (20 mm by 70 mm by panel thickness) were evaluated for resistance to mold fungi according to a modification of the ASTM D4445-10 (ASTM, 2010). Three mold fungi, *Aspergillus niger* 2.242, *Penicillium chrysogenum* PH02, and *Trichoderma viride* ATCC 20476 were grown and maintained on 2% malt agar (Difco, Detroit, MI, USA) at 27°C, and 80% RH. All fungi were obtained from the USDA Forest Service Forest Products Laboratory, Madison, WI, USA. A mixed spore suspension of the three test fungi were prepared by washing the surface of individual 2-week-old Petri plate cultures with 10-15 ml of sterile DI water. Washings were combined in a spray bottle and diluted to approximately 100 ml with DI water to yield approximately 3x10⁷ spores ml⁻¹. The spray bottle was adjusted to deliver 1 ml inoculum per spray. Specimens were sprayed with 1 ml of mixed mold spore suspension and incubated at 27°C and 80% RH for 12 weeks. Following incubation, specimens were visually rated on a scale of 0–5 with 0 indicating the specimen is completely free of mold growth and 5 indicating the specimen was completely covered with mold growth (0: no growth, 1: 20%, 2: 40%, 3: 60%, 4: 80%, 5: 100% coverage with mold fungi).

Thermal degradation and analysis

Thermal degradation of panels was carried out by using Perkin Elmer Diamond Thermal Analysis Instrument calibrated by using the melting points of indium (T_m=156.6°C) and tin (T_m=231.9°C) under the same conditions as the sample. The analyses of panels, kraft paper and LDPE were carried out at 15°C/min in an atmosphere of N₂ that had a constant flow rate of 100 ml/min. The samples (~5 mg) were allowed to settle in standard alumina crucibles and heated up to 700°C.

SEM investigations

Scanning electron microscopy examinations were performed by using a Zeiss EVO40 scanning electron microscope (Carl Zeiss SMT, Inc., Thornwood, NY). Elemental composition of cardboard panels was determined by energy-dispersive x-ray analysis using an IXRF Systems, Inc. (Houston, TX).

Results and Discussion

Figure 1 represents shredded particle from Tetra Pak cartons and panels manufactured in the study. Paper and aluminum parts of the waste material are easily recognized in both shredded particles and composite panels.



Figure 1. Shredded particles (a) and panel specimen manufactured (b)

Table 1 give mass losses in test specimens from panels in decay resistance tests. When compared to the mass losses occurred in Scots pine solid wood specimens served as controls, mass losses in panel specimens were considerably lower; however, panel specimens were not fully protected against the fungi. In the tests, *I. lacteus* resulted in higher mass losses in comparison with other test fungi.

Table 1. Decay resistance tests results obtained from panels

Decay type	Fungus	Mass loss (%)*
White-rot	<i>Irpex lacteus</i>	23.82 (2.56)
White-rot	<i>Coriolus versicolor</i>	13.30 (2.23)
Brown-rot	<i>Gloeophyllum trabeum</i>	15.22 (3.64)
Brown-rot	<i>Postia placenta</i>	15.94 (2.45)

*Values in parentheses are standard deviations. n:9. Mass losses in solid Scots pine wood specimens are: 47.03%, 40.95%, 56.06%, and 66.28% for *I. lacteus*, *C. versicolor*, *G. trabeum*, and *P. placenta*, respectively.

Termite resistance tests based on the JIS K 1571 standard test method (JIS, 2004) showed that panels had lower mass losses against *C. formosanus* termites when compared to solid Sugi wood specimens (Table 2). No termite mortality was seen in the panel specimens. Termite resistance test results based on the AWPA E1-06 standard test method (ASTM, 2010) using *R. flavipes* termites are given in Table 3. When compared to the JIS K 1571 test method using *C. formosanus* termites, much less mass losses were obtained in the panel specimens; however, termite mortalities of 13% were recorded in these tests. Termite resistance tests suggested that *C. formosanus* termites were more aggressive over the panel specimens based on the mass losses and termite mortalities occurred in the ASTM method by *R. flavipes* termites.

Table 2. Termite resistance test results (JIS K 1571)

Specimen #	Mass loss (%)			Mass loss (g)			Consumption rate ($\mu\text{g}/\text{day}/\text{termite}$)			Termite mortality (%)		
	Mean	Std Dev		Mean	Std Dev		Mean	Std Dev		Mean	Std Dev	
#1	7.91			0.21			60.02886			0		
#2	6.94			0.19			54.54545			0		
#3	7.87	7.57	0.55	0.21	0.20	0.01	60.31746	58.30	3.25	0	0.00	0.00

Mass loss in solid Sugi sapwood specimens is 27.87%.

Table 3. Termite resistance test results (AWPA E1-06)

Specimen #	Mass loss (g)			Mass loss (%)			Termite mortality (%)		
	Mean	Std Dev		Mean	Std Dev		Mean	Std Dev	
#1	0.13			1.14			12		
#2	0.31			2.87			15		
#3	0.00			0.00			10		
#4	0.06			0.54			12		
#5	0.12	0.12	0.24	1.12	1.13	2.23	15	12.80	2.17

Mass loss in solid Scots pine sapwood specimens is: 25.98%.

Mold resistance tests of the panel specimens revealed that the specimens were completely resistance against mold growth even at an extended period of 12 weeks (Table 4). Solid Scots pine sapwood specimens as controls were fully covered by the fungi at a 4-week-duration.

Table 4. Mold resistance test results

Specimen #	Week 4	Week 8	Week 12
#1	0	0	0
#2	0	0	0
#3	0	0	0
#4	0	0	0
#5	0	0	1
#6	0	0	0
#7	0	0	0
#8	0	0	0
#9	0	0	0
#10	0	0	1

Ratings: 0-5; 0: no mold growth, 5: complete coverage by mold fungi. Mold coverage in solid Scots pine sapwood specimens: 5

TG and DTG curves are given in Figure 2. It can be seen that the thermal degradation of panels can be divided into 3 phases after moisture evaporation. A comparison of the TG and DTG curves at a 15°C/min heating rate of panels, paper and LDPE are given in Figure 2. The panels are composed of three different components and their overall thermal degradation behavior can be explained by association of thermal properties of these components. Thermal degradation reactions of paper samples took place in two steps and began to occur at lower temperatures (<350°C) than those of both LDPE and panels. It can be seen that the thermal degradation of the LDPE layer, which took place in the main reaction, peaked at 432°C and the main reaction was followed by a smaller reaction (470°C). In addition, aluminum foil does not degrade during thermal treatment and melts at around 600°C. It seems that aluminum had no effect on the trend of TG and DTG profiles of the panels.

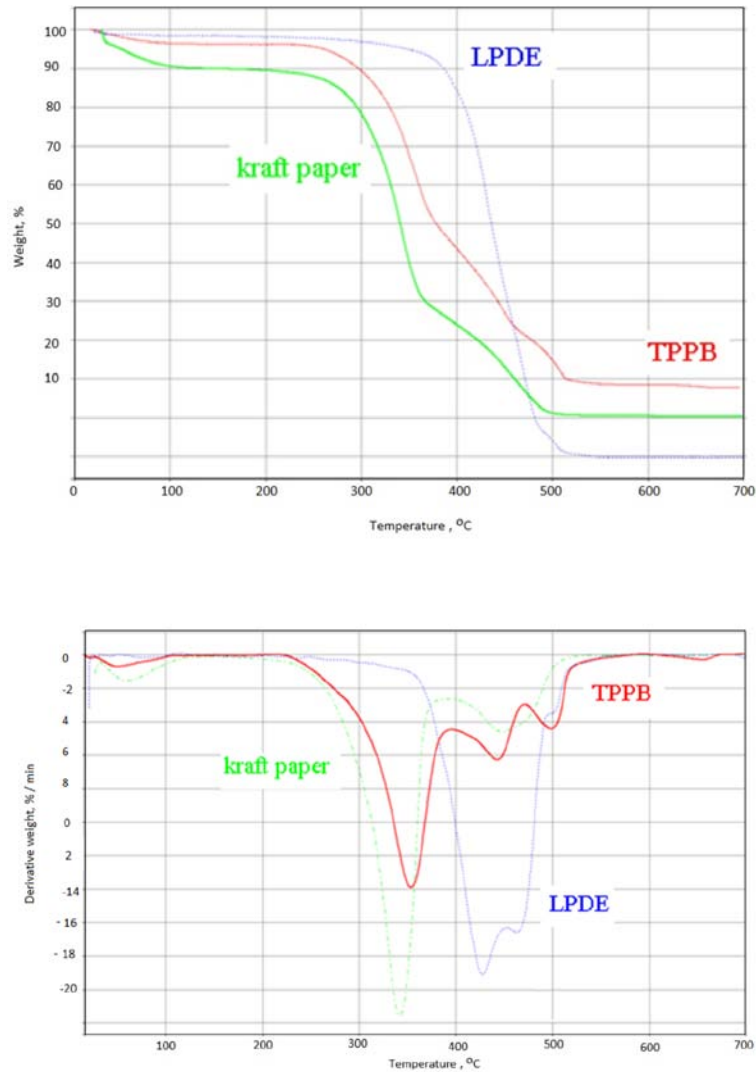


Figure 2. TG and DTG curves of panels (TPPB), kraft paper and LDPE

Distribution of paper, LDPE, and aluminum in the panels was evaluated by SEM micrographs (Figures 3 and 4). In the micrographs, while smooth areas indicate LDPE parts, relatively bright long strings represent aluminum. Cellulose fibers seem fractured and the figures obviously indicate decomposition of fibers. It is clearly seen that neither LDPE nor aluminum is chemically bonded with cellulose fibers. Therefore the mixture of plastic and fibers demonstrate non-uniform structure. The elemental distribution obtained from X-ray Microanalysis System and the semi-quantitative distribution of elements observed in the map of the panels shown in Table 5. Calcium (Ca) crystals were observed in SEM micrographs (Figure 4) and also determined by X-ray microanalysis system assuming that they are chemical coming from pulping process. The major disadvantage of cellulose fibers (about 75% of the panel structure) is low resistance to moisture. On the other hand, LDPE has a hydrophobic character and holds the structure together by melting.

Table 5. Distribution of elements on panel surfaces (%semi-quantitative)

C	O	Al	Si	Ca	Total (w/w)
78.0	17.5	3.6	0.2	0.7	100.00

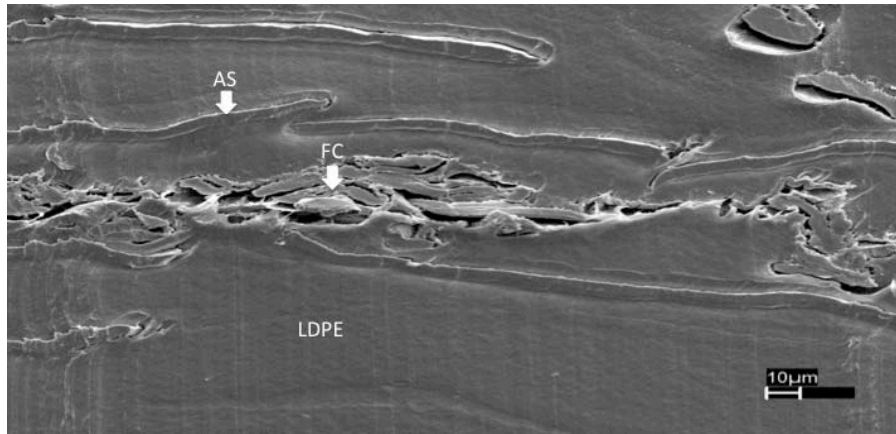


Figure 3. SEM micrograph of panels- cellulose fibers (CF), aluminum strips (AS) and LDPE

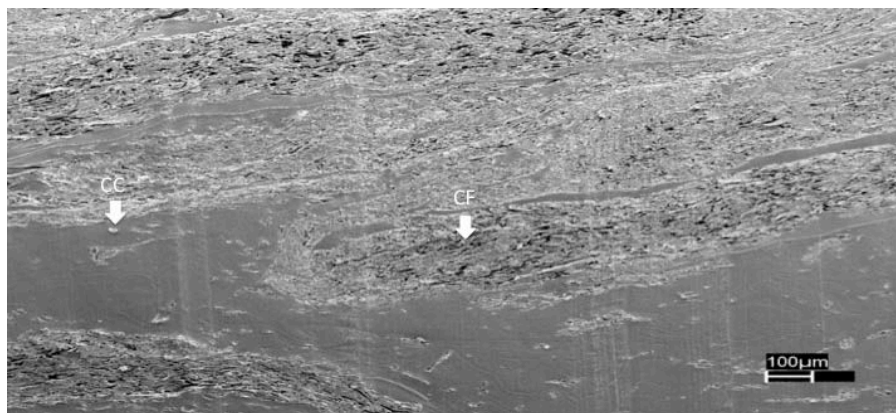


Figure 4. SEM micrograph of panels-distribution of cellulose fibers (CF) and Ca crystals (CC)

Conclusion

Biological performance tests reveal that panels without any biocide produced from waste Tetra Pak[®] cartons can be used for outdoor application where termite risks are available; however, such panels may be still susceptible to fungal decay. Indoor applications where no high humidity and no high moisture content are present, can be suitable for such panels without any biocide treatments based on mold resistance and termite resistance tests. For outdoor applications, biocides such as borates can be incorporated into such panels to increase biological resistance against fungal degradation. It is clear that panels with cellulose content of 75% lead to swelling and being susceptible to biological degradation and having weak dimensional stability. On the other hand, it is important to produce panels without adding any additives to reduce the costs and recovery of this kind of material without leaving any waste. Further studies are in progress to evaluate the effects of Zn-borate on biological and fire performance in order to increase biological performance of such panels when used outdoor purposes.

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