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A Laboratory Trial on the Usability of Waste Impregnated Papers in Standard Quality High Density Fiberboard (HDF) Production

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Research Article

Abstract− The aim of this study is that determination of the usability of the different types of waste impregnated papers (IP) in a standard HDF production line. For this purpose, the HDF panels were produced from industrial prepared fibers and waste of IP, called decor and overlay, preparation lines by three different ratios (from 5% to 15%). Some important properties of HDF's were determined according to standard test methods and were evaluated by statistical analysis in a 95% confidence interval. According to the evaluation results, it was concluded that it is possible to statistically evaluate the use of overlay and decor papers up to 15% in a standard production in terms of density, and equilibrium moisture, thickness swelling (2 h and 24 h), and modulus of rupture values. In addition, it was observed that the modulus of elasticity and tensile strength values generally improved as the amount of waste paper increased, regardless of the type of waste paper. However, the significant increase in formaldehyde emission showed that evaluating these waste papers without changing in the standard production line is difficult.

Keywords − *Impregnated papers, HDF, Mechanical properties, Physical properties, Formaldehyde emission*

Atık Emprenyeli Kağıtların Standart Kalite Yüksek Yoğunluklu Lif Levha (HDF) Üretiminde Kullanılabilirliği Üzerine Bir Laboratuvar Denemesi

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Araştırma Makalesi

Öz – Bu çalışmanın amacı, farklı türlerdeki emprenye edilmiş atık kağıtların (IP) standart bir HDF üretim hattında kullanılabilirliğinin belirlenmesidir. Bu amaçla endüstriyel olarak üretilen ve levha üretimine hazır liflere, dekor ve overlay olarak adlandırılan emprenye edilmiş kağıt atıklarından üç farklı oranda (%5'ten %15'e kadar) eklenerek HDF paneller üretilmiştir. Üretilen HDF'lerin bazı önemli özellikleri standart test yöntemlerine göre belirlenmiş ve %95 güven aralığında istatistiksel analiz ile değerlendirilmiştir. Değerlendirme sonuçlarına göre, standart bir üretimde %15'e kadar olan overlay ve dekor kağıtlarının kullanımının yoğunluk ve denge rutubeti, kalınlığı şişme (2 saat ve 24 saat) ve eğilme direnci değerleri açısından istatistiksel olarak değerlendirilmesinin mümkün olduğu sonucuna ulaşılmıştır. Ayrıca, atık emprenyeli kağıt türü fark etmeksizin, atık emprenyeli kağıt miktarı arttıkça elastikiyet modülü ve çekme dayanımı değerlerinin genel olarak iyileştiği gözlemlenmiştir. Fakat formaldehit emisyonundaki ciddi derecedeki artış, standart üretim hattında herhangi bir değişiklik yapılmadan bu atık kağıtların değerlendirilmesinin zor olduğunu göstermiştir.

Anahtar Kelimeler – *Emrenyeli kağıt, HDF, Mekanik özellikler, Fiziksel özellikler, Formaldehit emisyonu*

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1. Introduction

The majority of interior wood-based panels, such as particleboard (PB), medium-density fiberboard (MDF), and high-density fiberboard (HDF), are commonly utilized as surfaces with laminating applications (Dönmez Çavdar *et al*., 2013). Coating papers, such as decor and overlay papers, are one of these applications and are made of alpha-cellulose and impregnated with amino plastic thermosetting adhesives, such as melamine-formaldehyde (MF), urea-formaldehyde (UF), melamine-urea-formaldehyde (MUF), or some of them together (Ayrılmış, 2012). During the coating process at a MDF factory that consumes nearly 420 million m^2/year of impregnated paper (IP), almost 2.5 million m^2 of waste IP are generated (Başboğa *et al.*, 2016; Mengeloğlu *et al.*, 2016). Besides, it was known that 75% of the PB produced is laminated, and 96% of that is impregnated paper coated (Varga *et al.*, 2004). Impregnated decor paper wastes contain nearly 40% alpha cellulose, and 60% resin and additive chemicals (Alpár and Winkler, 2006; Başboğa *et al.*, 2018). However, Impregnated overlay paper wastes comprise 22% alpha-cellulose, 13% Al_2O_3, and 77.1% resin and their additive chemicals. Reusing or recycling these waste materials is the most effective way to reduce the amount of waste that ends up in landfills (Ayrılmış, 2012).

Today, many studies have been carried out to bring these wastes to industry instead of disposal. Varga *et al.* (2004) found that IP was applied to urea-formaldehyde as a mixture to improve particleboard's mechanical properties. Another study stated that powdered melamine impregnated paper (MIP) was used successfully as a binder for wooden chips (Le Fur *et al.*, 2004). Ayrılmış (2012) investigated the use of grinded waste MIPs as a binder in MDF. The result of this study was the same as Varga *et al.* (2004) study, but the authors stated that decorative paper had less positive effects than overlay papers. It was stated that it was possible to improve mechanical and physical properties by extending more than 40% of the waste MIPs in the oriented strand board (OSB) core layer (Dönmez Çavdar *et al.*, 2013). In another study, Mengeloğlu *et al.* (2016) tried to use waste MIP as a binder instead of UF in PB. Also, the authors stated that improvements were achieved in internal bond strength, bending strength and modulus of elasticity by using UF adhesives on the surface layers and MIP in the core layer. Another study found that the size of the particles and the ratio of the waste MIPs were important factors that affected the mechanical and physical properties of particleboards (Başboğa *et al.*, 2016). It was observed that waste IP provided worse results than the resin mixture, when waste MIP and an impregnation resin mixture were compared. However, some studies emphasized that the usability of IP as a connector was satisfactory (Başboğa *et al.*, 2018; Ramezanian Sani and Enayati, 2020).

HDF is used in many applications, such as workbenches, floors, and siding, which need thin thickness and high technological properties (Rowell, 2014). These desired high strength values in HDFs can be achieved with long fibers, but long fibers create a porous structure in the board mat, which results in the decline of some physical properties. Therefore, they are generally used as a mixture with short fibers in production (Park *et al.*, 2001). At this point, waste IPs can be used as filler instead of short fibers in the production of standard quality HDF. Because, all the studies on this topic show that the dimensional stability and mechanical properties of boards were improved by using waste IP; these wastes are also very suitable for HDF production to meet the desired properties. Although a significant amount of waste IP occurs, it is essential to use it within the quality control values in the HDF production line regarding standard quality product continuity. For this purpose, the focus of this study was to determine the usability as a filler material of the waste impregnated decor paper and waste impregnated overlay paper compared to other technological properties of HDFs without a statistically significant change in thickness swelling properties.

2. Material and Method

2.1. Materials

Industrially prepared wood fibers, obtained by the thermomechanical pulping method under factory conditions, were made from a softwood chip mixture of 70% fir and 30% pine wood chips. Wood fibers were glued with urea-formaldehyde resin (urea/formaldehyde molar ratio of 1/1, solid content of 56%, pH of 8.5, viscosity cPs of 20 s, density 1.240 gr/cm³, gelation time of 76 s) at 95 kg per cubic meter, and hardener (ammonium chloride 0.05 kg per cubic meter) and paraffin (20 kg per cubic meter) were added in the blow line. After the bunker before pre-pressure, the prepared softwood fiber mixtures were supplied from the fiberboard production line of the Kastamonu Entegre A.Ş. Kastamonu O.S.B. plant (Kastamonu, Türkiye). The waste décor and overlay paper wastes (Table 1) were provided by the impregnated paper lines of the Kastamonu Entegre A.Ş. Kastamonu O.S.B. plant (Kastamonu, Türkiye).

Table 1
Waste IP's properties

Type	Decor	Overlay
Weight (g/m^2)	125	95
Thickness (mm)	0.11	0.16
Alpha-cellulose (%)	38.4	20.9
Melamine-formaldehyde (%)	37.1	66.6
Urea-formaldehyde (%)	23.3	-
Additives (hardener,… etc.) (%)	1.2	1.0
Al_2O_3 (%)	-	11.5

2.2. Preparation of test samples

Wastes were milled in a hummer miller (2-2.5 mm width and 3-3.5 mm length) and mixed with prepared softwood fibers at different ratios of 5%, 10%, and 15% (Table 2). All HDF panels were manufactured with 32 kg/cm² at between 190-195 °C for 5 min at the target density of 850 kg/m3 with dimensions of 450 x 450 x 8.1 mm by a laboratory-scale single layer press at Kastamonu Entegre A.Ş. Kastamonu O.S.B. plant. Manufactured HDF panels were conditioned in ambient conditions (20±2 °C and 65±5% relative humidity) to reach a constant weight before determining the properties.

Table 2
Experimental design

Group name	Waste IP type	Waste IP Ratio
A	Decor	5%
B	Decor	10%
C	Decor	15%
D	Overlay	5%
E	Overlay	10%
F	Overlay	15%
G	-	-

2.3. Methods

Manufactured HDF panels' densities were determined by calculating the weight of the panels per unit volume according to EN 323 (1993) norms as eight replicates. The equilibrium moisture of constant-weight HDF samples was determined under EN 322 (1993) as four replicates. According to EN 317 (1993), twelve 50 x 50 x h mm samples for each group were put in water for 2 and 24 hours, and thickness swelling values were found by measuring the thickness of the pieces before and after water application. A universal testing machine (Imal IB 600 ®) performed a three-point loading test according to EN 310 (1993) with six sample replicates to determine the modulus of elasticity (MOE) and modulus of rupture (MOR). Internal bonding test samples were prepared using a silicone adhesive with 50x50 mm HDF panel samples bonded with the metal blocks. After tensile strength perpendicular to a surface test was performed for calculation of internal bond strength (IBS) for eight replicates using the universal testing equipment (Imal IB 600 ®) under EN 319 (1993). The formaldehyde release content was determined at two stages. Firstly the approximately, 100 g sample extracted by the perforator apparatus according to EN 120 (1992). After the extraction process, the formaldehyde content was analyzed photometrically using a LANGE LT 200 UV spectrophotometer in four replicates. The test results of the HDF samples were analyzed with the univariate variance test (ANOVA) and DUNCAN test by SPSS 23 (IBM Corp., Armonk, NY, USA) for statistical comparison between groups.

3. Results and Discussion

The average of obtained results of the physical properties was presented in Table 3. Table 4 shows the results of the ANOVA test to evaluate the effect of waste IP type and waste IP ratio. According to ANOVA results, physical properties were not significantly changed with the waste IP addition to the HDF panels increased to 15%. The fact that there are no statistically significant changes in the density and the thickness swelling values (2 h and 24 h), where the dimensional stability of the boards is measured, this situation is an indication that the waste IPs can be easily integrated into the production line in terms of production in uniform quality. Ramezanian Sani and Enayati (2020) reported that the waste MIP did not affect the thickness swelling value according to statistically. In addition, it has been proven in different studies in the literature that it is possible to add waste IP to the boards at higher rates if higher dimensional stability is desired, unlike providing a standard quality value (Ayrılmış, 2012; Başboğa *et al.*, 2018, 2023).

Table 3
Physical properties of high density fiberboards

Group	Density	Equilibrium moisture	Thickness swelling 2 h	Thickness swelling 24 h
A	860.85 (13.81)	7.39 (0.04)	5.59 (0.28)	12.27 (0.96)
B	866.12 (8.80)	7.18 (0.11)	5.46 (0.18)	11.65 (0.93)
C	869.67 (21.10)	7.09 (0.24)	5.34 (0.19)	11.53 (0.80)
D	863.23 (11.38)	7.05(0.12)	5.51 (0.31)	11.76 (1.22)
E	872.41 (7.57)	6.89(0.35)	5.44 (0.67)	11.70 (1.02)
F	870.20 (20.42)	7.17(0.11)	5.43 (0.45)	11.18 (0.92)
G	859.56 (7.99)	7.62(0.21)	5.70 (0.52)	12.56 (1.10)

Table 4
Variance analysis of physical properties of HDF panels

Property	Source	Sum of squares	df	Mean square	F	Sig.*
Density	Waste IP Type (A)	112.976	1	112.976	0.573	0.453
	Waste IP Ratio (B)	613.273	2	306.637	1.555	0.221
	A* B	69.138	2	34.569	0.175	0.840
	Error	9664.106	49	197.227		
	Total	42009128.117	56			
Equilibrium moisture	Waste IP Type (A)	0.131	1	0.131	3.483	0.076
	Waste IP Ratio (B)	0.009	2	0.004	0.120	0.888
	A* B	0.380	2	0.190	5.076	0.016
	Error	0.787	21	0.037		
	Total	1459.942	28			
2 h Thickness swelling	Waste IP Type (A)	4.737E-5	1	4.737E-5	0.000	0.986
	Waste IP Ratio (B)	0.349	2	0.174	1.129	0.329
	A* B	0.094	2	0.047	0.304	0.739
	Error	11.427	74	0.154		
	Total	2462.906	81			

Tablo 4 devam ediyor

24 h Thickness swelling	Waste IP Type (A)	1.255	1	1.255	1.261	0.265
	Waste IP Ratio (B)	5.241	2	2.621	2.633	0.079
	A* B	0.865	2	0.433	0.435	0.649
	Error	73.643	74	0.995		
	Total	11386.562	81			

*(p≤0.05) is significant

Regarding the mechanical properties (Table 5), ANOVA analysis (Table 6) revealed that waste IP type had no significant effect. However, the waste IP ratio significantly affected the MOE and IBS values. For this purpose, Figure 1, for MOE, and Figure 2, for IBS, were created by the DUNCAN test results to understand how waste IP ratio affects the type on panel values. In Table 5, although it is not statistically significant, it was investigated that the best MOR values were obtained from the overlay paper groups. The main reason for this can be explained by the fact that overlay papers contain more melamine formaldehyde than decor papers (Table 1). Because, melamine formaldehyde provides stronger bonds than urea-formaldehyde. As a result, overlay wastes provided a high level of wood fiber bonding (Ayrılmış, 2012). In addition, with the increase in waste paper content in general, mechanical properties also increased in parallel with the literature studies (Alpár and Winkler, 2006; Başboğa *et al.* 2016, 2018).

Table 5
Mechanical properties of high density fiberboards

Group	MOR	MOE	IBS
A	36.68 (2.79)	3468.74 (234.72)	1.29 (0.11)
B	38.70 (2.54)	3711.63 (276.47)	1.31 (0.09)
C	39.95 (1.53)	3779.33 (213.31)	1.37 (0.04)
D	39.22 (2.44)	3640.37 (234.45)	1.31 (0.04)
E	39.72 (0.75)	3650.87 (149.97)	1.32 (0.04)
F	40.35 (2.00)	3979.68 (208.93)	1.44 (0.12)
G	37.33 (2.59)	3586.13 (186.40)	1.18 (0.05)

Table 6
Variance analysis of mechanical properties of HDF panels

Property	Source	Sum of squares	df	Mean square	F	Sig.*
MOR	Waste IP Type (A)	15.629	1	15.629	3.235	0.081
	Waste IP Ratio (B)	29.115	2	14.558	3.013	0.062
	A* B	7.299	2	3.649	0.755	0.477
	Error	169.108	35	4.832		
	Total	63631.997	42			
MOE	Waste IP Type (A)	96861.001	1	96861.001	2.037	0.162
	Waste IP Ratio (B)	643807.295	2	321903.648	6.769	0.003
	A* B	123012.743	2	61506.372	1.293	0.287
	Error	1664426.487	35	47555.042		
	Total	573890985.605	42			

Tablo 6 devam ediyor

IBS	Waste IP Type (A)	0.013	1	0.013	2.203	0.144
	Waste IP Ratio (B)	0.096	2	0.048	8.112	0.001
	A* B	0.007	2	0.004	0.629	0.537
	Error	0.289	49	0.006		
	Total	97.452	56			

* ($p \leq 0.05$) is significant

It was clearly understood from Figure 1 that up to 10% waste IP can be used to produce standard quality production in terms of modulus of elasticity. The same figure shows that the mean values of the groups added with 15% are statistically significantly different and have a higher value compared to the control and 5% added groups. As a result, it can be easily said that waste IP could be evaluated in HDF production in terms of modulus of elasticity up to 10% in terms of a standard quality control.

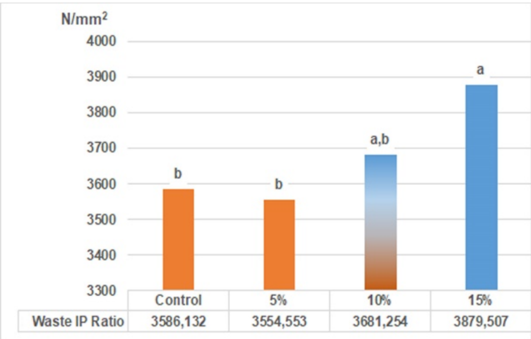

Figure 1 DUNCAN groups of the MOE values of HDF panels in terms of waste paper ratio

Figure 2 represented that the IBS values increased with increasing the waste IP ratio. The main reason is that the amount of adhesive in the composite board samples per unit area increases with the amount of the waste of IPs (Ramezanian Sani and Enayati, 2020). Generally, as the amount of adhesive increases in fiberboards, the IBS values increase (Hong *et al.*, 2017; Sıradağ *et al.*, 2018). Although the paper type was not statistically significant, Table 5 indicates that the overlay groups have higher values than decor papers because this paper type contains more adhesive than decor papers. When Figure 2 was examined, the control group had the lowest IBS value at 1.18 N/mm^2, while the average for the 15% waste IP including groups was 1.40 N/mm^2, which was nearly 19% higher than the control groups.

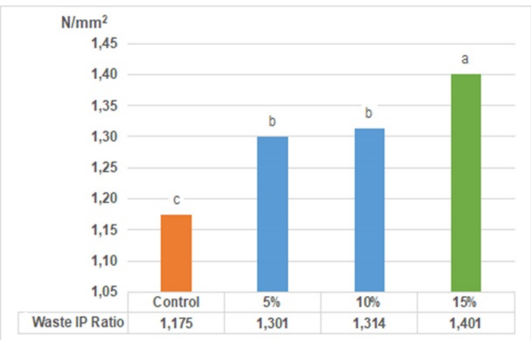

Figure 2 DUNCAN groups of the tensile strength values of HDF panels in terms of waste paper ratio

The most important disadvantage of fiberboards is formaldehyde emission, which arises from the formaldehyde based adhesives (such as urea formaldehyde, melamine formaldehyde and phenol formaldehyde) used and increases the risk of cancer with long-term exposure of the users (Mengeloğlu *et al.*, 2018). Figure 3 shows the formaldehyde emission values of the tested board samples. None of the groups' results met the E1 norm of standards (EN 622-1, (2003); EN 13986, (2004)). Formaldehyde emission values of the groups increased with the increase in the percentage of waste IP. The lowest value was obtained from the control group with 8.66 mg/100 gr. The highest value 13.40 mg/100 gr belongs to the 15% Overlay group and more than 54% of the control group values. It was thought that the main reason why the control samples did not meet the standards was that the fibers prepared according to the normal production conditions of the factory were produced under laboratory conditions. Because factors such as production temperature and pressure influence the amount of formaldehyde emissions from the boards (Istek *et al.*, 2018).

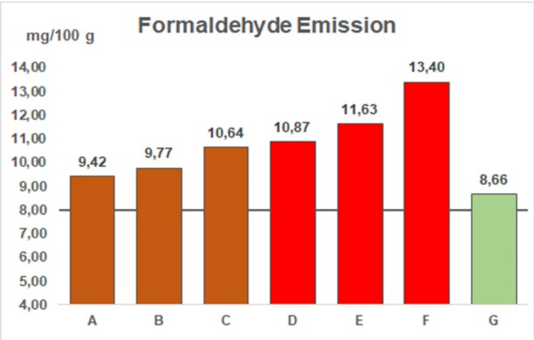

Figure 3 Formaldehyde emission values of tested panels

ANOVA test results were given in Table 7 to examine the effect of waste IP type and amount of waste IP on formaldehyde emission values. According to Table 7, the change of waste IP type and the ratio of used of waste IP changed, and the interaction effect of these two on formaldehyde emission values was found to be statistically significant ($p \leq 0.05$).

Table 7
Variance Analysis of Formaldehyde emission Properties of HDF panels

Source	Sum of Squares	df	Mean Square	F	Sig.*
Waste IP Type (A)	24.462	1	24.462	1609.230	0.000
Waste IP Ratio (B)	14.908	2	7.454	490.348	0.000
A* B	1.791	2	0.896	58.924	0.000
Error	0.319	21	0.015		
Total	3221.533	28			

∗ ($p \leq 0.05$) is significant

The results of the DUNCAN analysis performed according to the results of the ANOVA analysis were given in Figure 4. It was observed from Figure 4a that there was a significant difference in the 95% confidence interval between the control group, the waste decor paper added groups, and the waste overlay paper added groups in the samples' formaldehyde emission test. The waste overlay paper added groups have higher formaldehyde emission values than the waste decor paper added groups. The main reason for this can be shown by the fact that overlay paper wastes contain more formaldehyde based adhesive than decor paper wastes, according to Table 1. In addition, in Figure 4b, an increase in formaldehyde emissions was observed with the increase in the amount of waste IP at the 95% confidence interval. The main cause of this situation is an increase in the amount of glue in the fiberboards produced as the amount of waste IP increases. Also, an increase in formaldehyde emission values is an expected situation for boards with waste IP, and this is the case in many literature studies (Alpár and Winkleri, 2006; Başboğa *et al.*, 2018, 2023). Such an increase in formaldehyde emissions is a major issue that must be addressed to maintain standard production practices. In order to solve this problem, different formaldehyde scavengers, such as amine-based chemicals, should be applied, as in the study of Mengeloğlu *et al.* (2018).

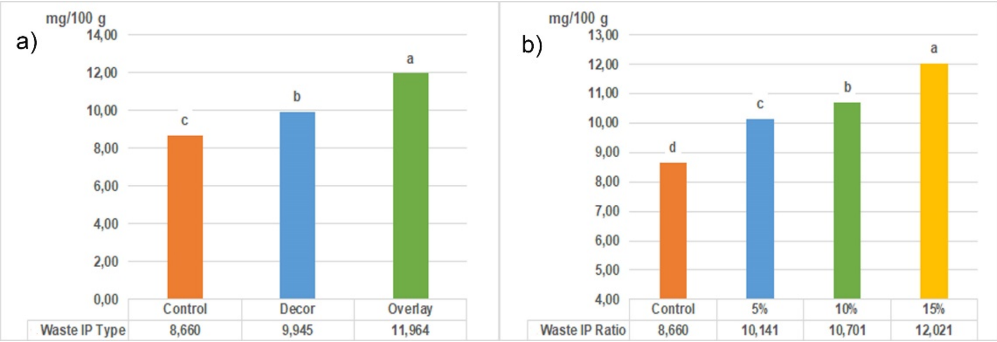

Figure 4 Duncan Analysis Results of Formaldehyde Emission values a) waste IP type b) waste IP ratio

4. Conclusion

Briefly, the following conclusions can be concluded from this study:

- It has been observed that waste IPs can be used in standard HDF production up to 15%, since IPs do not statistically affect thickness swelling (2h and 24 h), density, and equilibrium moisture values.
- All parameters tested in this study can be accepted for a standard HDF production in terms of modulus of rupture according to the variance analysis tested in this study.
- As a result of MOE values, up to 10% of waste IP can be evaluated to produce a standard quality HDF.
- The IBS of HDF improves with the amount of waste IP into the fiberboard, regardless of the waste IP type.
- The formaldehyde emission values of HDF samples increase due to the intense formaldehyde-based adhesives in waste IPs. This situation is a major disadvantage for evaluating the IPs' in the HDF production line. Future studies should focus on reducing formaldehyde emissions in different ways.

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Author Contributions

Nazif Özgen KUTLUATA: carried out experiments, data collection, and reporting.
Çağrı OLGUN: analyzed the data, drafted the article and revisions
Mahmut GÜR: designed and planned the analysis.

Conflict of Interest

No potential conflict of interest was reported by the authors.

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