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Determination of Some Technological Properties and Formaldehyde Gas Release in Lignocellulosic Based Interior Design Panels

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Abstract

Aim of study: The aim is to produce lignocellulosic-based interior design panels (LIDP), a type of composite that complies with gas emission standards, from annual facility waste.

Material and method: The wood chips of Turkish pine and annual plant wastes cotton stalks, sunflower stalks and wheat stalks were used as test materials. Within the scope of the tests, the water uptake (WU) and thickness increase (TI) values were measured from physical properties and some technological properties (Janka hardness (JH), surface vertical screw holding strength (VSH), nail holding strenght (NHS) and surface soundness strength (SSS)) awere measured. Similarly, the FGR values of the LIDP were determined based on perforator method.

Main results: The measurement results of WU and TI in physical properties showed that the test panels did not meet the criteria required in the usage standards. In terms of technological features, as the annual waste plant ratio in the panel matrix increased, the power values also decreased. The best results in formaldehyde gas emissions evaluated according to EN 13986 were obtained in LIDP produced from WCR-SSR mixtures.

Research highlights: Investigation of the suitability of annual plant residues for the production of environmentally friendly lignocellulosic-based interior design panels (LIDP) with standard features.

Keywords: Composite Panel, Urea Formaldehyde, Physical and Technological Properties, Formaldehyde Gas Release

Lignoselülozik Esaslı İç Mekân Tasarım Panellerinde Bazı Teknolojik Özelliklerin ve Formaldehit Gazı Salınımının Belirlenmesi

Öz

Çalışmanın amacı: Yıllık tesis atıklarından gaz emisyon standartlarına uygun bir kompozit türü olan lignoselülozik esaslı iç tasarım panelleri (LIDP) üretmektir.

Materyal ve yöntem: Test materyali olarak kızılçam yongaları ve yıllık bitki artıklarından pamuk sapları, ayçiçeği sapları ve buğday sapları kullanıldı. Testler kapsamında fiziksel özelliklerden su alma (WU) ve kalınlık artış (TI) değerleri ölçülmüş ve teknolojik özelliklerden bazıları (Janka sertliği (JH), yüzeye dik vida tutma mukavemeti (VHS), çivi tutma mukavemeti (NHS) ve yüzey sağlamlık mukavemeti (SSS)) ölçüldü. Benzer şekilde kompozit panellerin formaldehit gaz salınım (FGR) değerleri perforator yöntemine göre belirlendi.

Temel sonuçlar: Fiziksel özelliklerde su alma ve kalınlık artışı ölçüm sonuçları, test panellerinin kullanım standartlarında aranan kriterleri karşılamadığını gösterdi. Teknolojik özellikler açısından panel matrisindeki yıllık atık tesis oranı arttıkça direnç değerleri de azaldı. EN 13986'ya gore değerlendirilen formaldehit gazı salınımlarında (FGR) en iyi sonuçlar WCR-SSR karışımlarından üretilen LIDP'de elde edildi.

Araştırma vurguları: Yıllık bitki artıklarının standart özelliklere sahip çevre dostu lignoselülozik esaslı içmekân tasarım panelleri (LIDP) üretimine uygunluğunun araştırılması.

Anahtar Kelimeler: Kompozit (LIDP), Üre Formaldehit, Fiziksel ve Teknolojik Özellikler, Formaldehit Gazı Salınımı

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Introduction

Since forest resources are limited and scarce, researching the possibilities of using alternative raw material resources in the forest industry has become important in recent years. Because difficulties in supplying raw materials cause production costs to increase. The protection and sustainable use of forests is becoming increasingly important due to increasing demand. Recycled forest products from lignocellulosic residues can be an important solution to meet these demands. Such recycling plays a critical role in preventing the depletion of natural resources and ensuring environmental sustainability. Such innovative approaches can contribute to economic growth while ensuring the preservation of natural forests (Youngquist et al., 1993; Bektas et al., 2005). Deforestation not only affects wood supply, but also leads to biodiversity loss, climate change and the degradation of local ecosystems. Forests play a critical role in combating climate change by absorbing carbon dioxide. Such deforestation can also cause economic, environmental and social problems in developing countries (Pirayesh & Khazaeian, 2012). The increase in demand for composite wood products in recent years is quite remarkable. These products are generally preferred due to their durability, environmental friendliness and the variety of aesthetic options they offer (Ashori & Nourbakhsh, 2008; Kim, 2009).

Composite panels are materials formed by turning residue such as wood sawdust, sawmill sawdust, wood residues, agriculrural residues and even sawdust into panels under a certain tempeture and pressure with the help of a synthetic resin or a suitable adhesive. Composite panels are a very popular wood-based panel product used in furniture cabinets, subflooring, home construction, doors, dining tables, safes and sports equipment, countertops, kitchen frames, wardrobes, joinery, pool edges, garden terraces, balconies, exterior cladding, indoor and outdoor applications, children's playgrounds, pergolas and many other areas (Rokiah et al., 1987; Bardak et al., 2011).

Formaldehyde is a chemical compound that is widely used, especially in the wood processing industry, and is often used in conjunction with binders. Urea formaldehyde (UF) resins are widely used in the panel industry, especially in the production of particleboard (such as OSB, MDF) and chipboard. UF resins are less costly than other binders. They also offer an efficient solution with fast drying times and strong binding capacity in the production process (Kim et al., 2006). However, Formaldehyde emissions can cause serious health problems. Long-term exposure in particular can increase the risk of respiratory diseases, allergic reactions and cancer (Lebkowska et al., 2017).

According to statistical studies, the amount of UF resin adhesive constitutes 91% of wood-based panel adhesives (Gu, 2015). However, composite panels produced using UF have the potential to release free formaldehyde, posing a serious threat to human health. Many scientists are working to reduce formaldehyde emissions from wood-based panels (Hashida et al., 2006). However, most of these methods shave high costs or secondary pollution problems (Ghani et al., 2018).

Nowadays, there is a need for alternative sources to replace wood raw material. Agricultural residues can play a leading role in maintaining the balance between supply and demand in the production of composite panels such as chipboards (Nemli & Aydin, 2007). These composite sheets are generally produced from agricultural residues such as walnut (Guru et al., 2008), sunflower stalks (Bektas et al., 2005), chestnut bur (Liang et al., 2021), kiwi prunings (Nemli, 2003), cottonseed husks (Gurjar, 1993), rice straw tree (Yang &Kim, 2003), linseed knife (Papadopoulos & Hague, 2003), vine prunings (Ntalos & Grigoriu, 2002), walnuts, pine cones (Buyuksari et al., 2010), almond shells (Hamidreza et al., 2013; Guru et al., 2006), wood flour (Kamdem, 2004), sugar cane bagasse and castor oil (Fiorelli et al., 2013), coffee husks and shells (Bekalo & Reinhardt, 2010), agricultural residues (Ferraz et al., 2020) and peanut residues (Gatani et al., 2013). The annual amount of waste in Turkey is approximately 142,4 million tons year^{-1} (Saka & Yılmaz, 2017). Guler (2015) emphasized that the amount that can be collected from these wastes is 37 million tons.

The aim of this study is to produce lignocellulosic-based interior design panels (LIDP) in accordance with gas emission standards from annual facility wastes, which are known as environmentally friendly raw material sources. At the same time, determining some technological properties and gas (formaldehyde) release values of the products to be manufactured are also among the objectives of the study.

Material and Methods

All plant and woody raw materials used in the study were supplied from the Eastern Mediterranean Region and include cotton (*Gossypium hirsutum L.*), sunflower (*Helianthus annuus L.*) and wheat (*Triticum aestivum L.*) stems and red pine (*Pinus brutia Ten.*) wood chips.

Lignocellulosic-based interior design panels (LIDP) produced using standard procedures under laboratory conditions require a meticulous process. Annual plant wastes and wood chips were first cleaned and then shredded by a double-blade chipper. Then, shredded annual plant waste and wood chips were classified on a laboratory classification sieve. Chips ready for production were dried in a drying oven at 100 ± 3 ºC to reach the desired moisture content (3%). 10% urea formaldehyde (UF) resin was used in the production of single-layer composite panels. In the panels produced as a single layer, 10% of the dry chip weight was added as a completely dry glue and 1% hardener (NH_4Cl) was added in proportion to the dry chip weight. The chips were placed in a drum mixer and sprayed with urea formaldehyde and ammonium chloride for 4 minutes to obtain a homogenized mixture. Thickness of LIDP was controlled by stop bars. Three composite panels were produced for each group. The dimensions of the produced composite panels were 50x50x1.8 cm (length x width x thickness) and panels target density was 650 g cm^{-3}. The produced LIDP conditioned at 23 ± 2 ºC and 65 ± 5% relative humidity to reach moisture content of about 12% before trimming to final dimension of 50x50x1.8 cm (length x width x thickness). Production parameters of composite panels were also displayed in Table 1.

Then, it was kept in the air conditioning cabinet at 20 ± 2 C and relative humidity of 65 ± 5% for 21 days and acclimatized according to the standard required in TS 642-ISO 554. (Bektas et al., 2005; Güler & Sancar, 2017). The production parameters given in Table 1 were applied for all LIDP types. Figure 1 shows the preparation way of LIDP boards.

Some technological properties janka hardness (JH) EN 2479 (1976), surface vertical screw holding strength (VSH) EN 320 (2011), nail holding strenght (NHS) American Society for Testing and Materials - ASTM D 1761 (2005) and surface soundness strength (SSS) EN 311 (2005) and WU and TI values were measured from physical properties according to EN 317 (1993).

Table 1. Production parameters of LIDP

Parameters	Values	Parameters	Values
Press temperature(ºC)	185	Thickness (mm)	18
Pressing time (min)	7	Dimensions (mm)	50x50
Pressure (kg mm^{-2})	20	33% NH_4CI content(%)	1

Random samples were taken from each panel type to determine formaldehyde emissions using the perforator method based on the EN 120 (1996) standard included in the BS 13986 standard. Analysis of variance (ANOVA) and Duncan test were applied at 95% confidence level to reveal statistical differences in the physical properties of particleboards.

In this method, 25x25 mm samples with moisture determination were weighed by calculating that 100 g would be completely dry. These weighed samples were placed in a 1000 mL glass flask and 600 mL of pure toluene was added. The samples were boiled in toluene for 2 hours. Then, the separated formaldehyde was allowed to pass into distilled water.

At the end of this period, the solution was extracted and then cooled to bring its volume to a certain level. In this case, distilled water was added to complete the volume of the solution. The goal in this process is to eliminate the effect of the solvent and obtain a certain density or concentration. In the last step, the formaldehyde in the solution was determined photo metrically with the help of a spectrophotometer. The experimental design is shown in Table 2.

Table 2. Sample production compositions

Board Type	Raw materials and mixing ratios (%)												
	P1*	P2	P3	P4	P5	P6	P7	P8	P9	P10	P11	P12	P13
WCR	100	75	50	25	0	75	50	25	0	75	50	25	0
CSR	0	25	50	75	100	0	0	0	0	0	0	0	0
SSR	0	0	0	0	0	25	50	75	100	0	0	0	0
WSR	0	0	0	0	0	0	0	0	0	25	50	75	100

*Control group, WCR: Wood chips ratio; CSR: cotton stalk ratio, SSR: sunflower stalk ratio, WSR: wheat stalk ratio

(a) (b) (c) (d)

Figure 1. Production stages of test samples: a)-In the field, b)-Chipping, c)-Pressing, d)-Sample

Results and Discussion

Results of ANOVA and Duncan's mean separation tests WU and TI values of the composite panel made using annual plant and wood sawdust mixtures with water immersion times of 1, 3, 48, 72, 96 and 336 hours are shown in Table 3. It is noted that Table 3, TI and WU values also increased with the increase in the use of annual plant wastes in LIDP. According to the TS EN 312 (2012) standard, the maximum TI value of particleboard after soaking in water for 2 hours must be 8%. Furthermore, according to the EN 312-4 (2012) standard, the maximum TI requirement after soaking in water for 24 hours is 15%. Wood contains a large number of free –OH groups. A hydroxyl group is a functional group with the chemical formula –OH. The hydroxyl group usually helps to make a molecule more polar and better able to dissolve in water. (Gwon et al., 2010; Nourbakhsh et al., 2011).

Retention of water within the composite structure may cause various structural and functional problems such as deterioration in mechanical properties (Ashori & Nourbakhsh, 2010). Hydroxyl groups can affect the biological and mechanical properties of holocellulose. These groups can change the water sensitivity of the polymers. Together with cellulose and hemicellulose, lignin strengthens the cell walls of plants and increases the resistance of plants to water. Extractives are substances found in plants that are extracted from plant material, usually with organic solvents (e.g. alcohol, ether, water). These substances cannot absorb water. (Gwon et al., 2010).

The increase in water uptake with increasing proportion of annual waste plants in the mixture matrix can be attributed to the presence of less extractive substances and lower holocellulose content in these materials than wood, as discussed above. The positive role of extractive substances on water resistance has been expressed by many researchers (Dunk & Pizzi 2002; Ayrilmiş et al., 2009; Buyuksari et al., 2010). Within the specified waiting periods, the maximum water uptake value was obtained in group P13 boards produced from 100% wheat stalks (156.54%), and the minimum WU value was obtained in group P1 boards produced from 100% woodchips (63.29%).

Since the density of annual plant stems is low and their permeability is high, the amount of water uptake is higher in the boards with less density.

Table 3. Statistical analysis results of water uptake and thickness increasetests(*)

Board type	ST (h)	Water uptake (WU)			Thickness increase (TI)		
		Mean (%)	Standard deviation	COV (%)	Mean (%)	Standard devision	COV (%)
P1	1	63.29a(**)	16.98	26.82	19.61a	14.75	79.25
P2		64.59a	16.98	93.56	22.21a	15.27	24.84
P3		67.49a	19.44	91.52	23.66a	16.64	73.45
P4		68.4a	13.13	83.99	24.23a	19.50	83.96
P5		71.06a	23.99	87.45	26.14a	35.82	142.44
P6		65.39a	26.64	14.44	21.21a	1627	24.84
P7		67.98a	13.13	19.31	22.66a	16.64	73.45
P8		72.69a	16.10	46.76	23.23a	19.50	83.96
P9		75.21a	18.34	48.72	25.14a	35.82	142.44
P10		95.02b	12.76	29.40	26.21b	16.27	24.84
P11		100.92b	28.13	35.12	29.66bc	16.64	73.45
P12		107.83b	23.52	49.34	33.23bc	19.50	83.96
P13		114.90b	22.28	42.02	35.14bc	35.82	142.44
P1	3	71.64a	24.22	19.85	22.02a	18.33	91.53
P2		74.3a	21.82	56.65	24.20ab	15.34	66.12
P3		76.87a	22.21	88.22	25.60ab	19.44	38.37
P4		77.1a	19.31	56.55	26.89ab	13.00	50.22
P5		81.14a	22.31	89.1	28.93b	22.21	39.44
P6		78.90ab	19.13	1152	23.20ab	15.34	66.12
P7		85.43b	15.61	18.27	24.60ab	19.44	38.37
P8		86.33b	16.36	53.70	25.89ab	13.00	50.22
P9		89.33b	24.66	27.60	27.93b	11.12b	39.44
P10		103.92b	12.69	31.46	33.20ab	15.34	66.12
P11		109.28bc	18.36	25.95	36.60ab	19.44	38.37
P12		120.10bc	25.76	38.10	40.89ab	13.00	50.22
P13		125.36c	15.62	28.41	44.93b	11.22	39.44
P1	48	98.35a	20.94	17.31	25.44a	24.71	98.48
P2		100.12a	18.84	45.11	29.21a	15.75	20.38
P3		103.1a	12.39	72.47	31.32a	18.58	61.26
P4		105.48a	22.38	30.86	33.09a	18.90	58.88
P5		106.82a	21.53	61.24	34.62a	19.12	56.83
P6		100.00a	15.28	15.28	28.21a	16.75	20.38
P7		103.88a	14.89	33.59	30.32a	18.58	61.26
P8		105.96a	18.97	36.78	32.09a	18.90	58.88
P9		108.20a	24.19	22.35	33.62a	19.11	56.83
P10		131.26b	14.70	26.44	38.21b	15.75	20.38
P11		134.74b	15.45	33.73	42.32bc	18.58	61.26
P12		138.48b	15.25	32.68	44.09bc	18.90	58.88
P13		142.89b	19.70	27.79	49.62ac	19.14	56.83
P1	72	104.67a	17.04	16.28	28.35a	17.28	63.19
P2		106.39a	26.92	25.31	30.11a	15.45	17.52
P3		110.18a	25.19	68.24	32.95a	19.14	59.91
P4		113.09a	18.69	25.37	34.78a	15.83	46.86
P5		115.84a	11.54	70.39	36.23a	16.34	46.37
P6		105.57a	20.21	18.97	29.11a	15.16	17.52
P7		110.17a	16.61	33.23	31.95a	19.14	59.91
P8		114.79a	18.73	33.74	33.78a	15.83	46.86
P9		116.85a	11.77	27.19	35.23a	16.34	46.37
P10		134.42b	18.17	20.95	43.11b	11.51	17.52
P11		136.36b	12.60	31.24	48.95bc	19.14	59.91
P12		140.09b	16.95	26.37	52.78bc	15.83	46.86
P13		147.54b	16.62	24.82	55.23c	16.34	46.37

Table 3. (Continued)

Board type	ST (h)	Water uptake (WU) Mean (%)	Standard deviation	COV (%)	Thickness increase (TI) Mean (%)	Standard devision	COV (%)
P1	96	108.49a	16.71	15.41	30.25a	17.71	58.48
P2		110.35a	17.66	34.13	32.58a	14.56	14.44
P3		114.22a	20.19	54.45	34.73a	18.45	54.69
P4		117.7a	13.57	28.52	35.43a	18.85	25.72
P5		119.29a	18.44	65.76	37.87a	14.54	39.45
P6		109.85a	17.66	34.13	31.58a	14.56	14.44
P7		111.16a	20.19	61.45	33.73a	18.45	54.69
P8		117.27a	13.57	28.52	34.43a	18.85	25.72
P9		120.73a	18.44	65.76	36.87a	14.54	39.45
P10		139.70b	11.47	22.53	45.11b	14.56	14.44
P11		140.03b	10.19	28.70	50.95bc	18.45	54.69
P12		146.19b	15.08	23.99	54.78bc	18.85	25.72
P13		151.33b	18.81	19.33	56.23c	14.54	39.45
P1	336	112.77a	16.29	18.17	35.77a	19.16	29.23
P2		116.5a	22.82	19.61	36.56a	13.47	32.26
P3		119.52a	20.11	58.57	37.83a	16.28	44.20
P4		120.14a	16.51	30.39	38.09a	16.99	45.80
P5		133.4a	17.86	33.35	43.18a	14.79	28.64
P6		115.23a	12.84	19.61	35.56a	15.47	32.26
P7		118.19ab	10.23	58.57	36.83a	16.28	44.20
P8		131.02b	16.51	30.39	37.09a	16.99	45.80
P9		132.70b	17.86	14.35	41.18a	15.79	28.64
P10		148.99b	15.98	30.86	52.11bc	13.47	32.26
P11		151.22b	13.21	35.18	54.95bc	16.28	44.20
P12		152.84b	12.44	19.92	56.78bc	16.99	45.80
P13		156.54b	14.83	22.25	58.23c	14.79	28.64

(*)The number of samples is 30, STSoaking time, COVCoefficient of variation, (**)According to Duncan's multiple comparison test (P>0.05), there is no statistically significant difference between the mean values indicated by the same letters.

By increasing the density of the boards, diffusion became difficult and there was a decrease in the amount of uptake. In the literature, as the density increases, the swelling rate decreases as a result of short-term immersion in water, due to the difficulty of water penetration. On the other hand, all other physical and mechanical properties increase. (Akbulut, 1991). In the literature, It has been observed that the amount of water uptake in boards made from sunflower, tobacco stalk and tea factory waste is 37-48% for 2 h, 60-71% for 24 h, the thickness increase is 17-29% for 2 hours, 22-37% for 24 hours (Kalaycıoglu, 1992).

It is stated that the thickness increase in flax stalk sheets is 20% and in hemp it is 25% (Kozlowski et al., 1987). According to these studies, it can be said that the TI and WU amount of sunflower and cotton stalk particleboards are higher than the standard values, but are close to the literature values.

The average TI and WU values of all boards' types showed not significant difference (*P>0.05*) from each other in TI and WU after 1, 3, 48, 72, 96 and 336 hours water immersion times.

This situation was also confirmed by the Duncan test results of the variance sources averages given in the Table 3. The amount of WU increased due to the increase in the annual plant waste rate from 0% to 100% and the extension of the soaking time.

When Table 3 above is examined, with the increase in the participation rate of cotton stalk, sunflower stalk and wheat stalk used in the production of test boards, the amount of increase in thickness also increased in all holding periods. Within the applied water absorption periods, the highest amount of TI was calculated in group P13 boards produced from 100% wheat stalks (58.23%), and the lowest amount of TI was determined in group

P1 board produced from 100% wood chips (18.61%).

As it is known, the increase in thickness and water uptake values of chipboards directly depend on the variables in production. The density of the boards affects the amount of increase in thickness of the board. The amount of increase in thickness is small in boards with high density, and relatively higher in boards with low density. It is stated in the literature that there is a decrease in the amount of water uptake with increasing density (Kalaycioglu & Colakoglu, 1994). According to the results of variance analysis, it was seen that the difference between the groups was not significant (P>0.05). Similar data were obtained in the Duncan test results of the variance sources averages (Table 3). However, it is seen that the measured thickness increase values of the boards produced in the study do not meet the mentioned limits of the standard.

Previous studies also indicated that the addition of annual plants in composite boards increases the thickness increment values of the produced boards compared to the wood (Yasar & Icel, 2016). In composites produced from sunflower stalks, it was revealed that the TI values of the samples changed depending on the sunflower concentration in the board (Bektas et al., 2005). Similarly, Kozlowski and Piotrowski (1987) reached 20% TI values in composite boards obtained from flax stalks and 25% TI values in composite boards obtained from hemp stalks.

The water uptake and thickness increase increment results, including control samples, couldn't meet the minimum requirements required in the EN 312 (2005) for general purpose use and interior equipment. In order to eliminate this negativity, the use of paraffin is recommended by Ugur (2021).

Table 4 shows the results of ANOVA and Duncan's mean separation analysis applied to the measured data for technological properties and formaldehyde gazrelease (FGR). It can be seen that all of the ANOVA analysis results applied to the technological properties and FGR values in Table 5 were at significance levels ($P<0.05$). At the same times, the test results measured on the technological features of the test boards produced from annual plants and their mixtures were lower than the control group samples.

The strength values of the test samples in Table 4 are listed as follows: The highest strength values were obtained from Cotton Stalks (P2, P3, P4, P5), while the lowest strength values were obtained from Wheat Stalks (P10, P11, P12, P013). In addition, from the Duncan's mean separation test results in Table 4, it can be said that annual plant species and additive ratios in technological properties create significant differences between resistance values in general. As a result, the technological properties of LIDP produced from agricultural waste can be improved by additional processing; chemical modification of the particle (Ndazi & Tesha, 2006; Abdolzadeh et al., 2011), biological treatments of anaerobic digestion (AD) (Zheng et al., 2009), use of Methylene diphenyl diisocyanate (MDI) resin instead of UF (Yang & Zhang, 2004), surface coating of the final product (Nemli et al., 2003), and use of nanoparticles to improve mechanical properties, use of bio-based composites (Lei et al., 2008; Roumeli et al., 2010).

Similar positive results are also achieved for panels made with agricultural wastes and unused raw materials (Ashori & Nourbakhsh, 2008; Guntekin & Karakus, 2008; Nemli et al., 2009; Ayrilmis et al., 2009; Buyuksari et al., 2010; Tabarsa et al., 2010).

When the technological properties in Table 4 are compared with the chemical components (cellulose, hemicellulose and lignin) given in Table 5, according to the annual plant wastes mixing ratios (25%, 50%, 75% and 100%), the ascending sort of strength values in question will be seen to confirm the effect of cellulose and lignin mentioned above. While high lignin content increases the physical durability of the plant, high cellulose content decreases the physical durability of the plant (Papadopoulos & Hague, 2003).

The lowest resistance values in all mixing ratios were obtained in WSR with the lowest cellulose percentage (33.78%). The effect of cellulose and lignin ratios is particularly evident on NHS, JH and VSH. The mechanical test results of the CSR boards (P2, P3, P4, P5) met the minimum criteria required for general-purpose use and interior fittings, including furniture manufacturing, in the EN 312 (2005), except for SSS and JH.

The status that the technological properties of the panels produced from sunflower stalks (P6, P7, P8, P9) meet the criteria required in the EN 312 (2005) are exactly the same as those expressed for CSR panels. As for WSR boards (P10, P11, P12, P13), the rate of the results obtained from them to meeting the EN 312 (2005) is quite different from the results of CSR and SSR panels.

Table 4. Analysis results of technological properties and formaldehyde gas release

BT	SV	NHS (MPa)	JH (MPa)	VSH (MPa)	SSS (MPa)	FGR (%)
P1	Mean	584.8c(*)	54.41c	1089.4a	0.68a	3.53a
	SD	47.39	8,84	217.2	0.04	0.13
	COV (%)	25.20	16.25	19.93	17.84	39.11
P2	Mean	551.5bc	50.68c	1018.8ab	0.58b	3.64a
	SD	126.01	16.25	212.5	0.01	1.00
	COV (%)	22.85	23.27	20.50	13.52	13.64
P3	Mean	524.8abc	42.92b	958.8bc	0.54	6.28abcd
	SD	121.01	8.62	154.1	0.04	2.44
	COV (%)	23.17	20.28	16.36	10.11	17.64
P4	Mean	471.5ab	40.54b	935.0bc	0.51b	8.63cd
	SD	101.90	7.18	110.9	0.02	81.52
	COV (%)	21.61	17.70	12.33	18.71	27.70
P5	Mean	447.7a	32.92a	908.1c	0.50b	9.26d
	SD	90.78	2.71	83.3	0.02	2.59
	COV (%)	20.68	8.24	9.35)	12.57	36.63
P6	Mean	569.9c	31.41a	891.7cd	0.39c	3.90a
	SD	147.4	8.34	100.3	0.11	0.29
	COV (%)	25.20	26.56	11.24	28.37	7.38
P7	Mean	531.7bc	30.44a	883.1cd	0.54b	4.84ab
	SD	101.0	9.03	69.7	0.05	0.43
	COV (%)	19.13	29.65	7.90	8.70	7.19
P8	Mean	474.9ab	28.52a	817.0de	0.57b	6.28abcd
	SD	64.1	3.78	128.5	0.02	2.45
	COV (%)	13.49	13.24	15.73	2.65	8.91
P9	Mean	437.5a	27.84a	792.2e	0.68a	7.19bcd
	SD	61.12	4.03	83.6	0.02	1.27
	COV (%)	13.96	14.49	10.55	2.94	17.61
P10	Mean	462.9c	29.31a	753.3ef	0.38c	5.10ab
	SD	156.1	14.57	15.78	0.02	0.27
	COV (%)	27.29	50.19	15.78	5.11	10.44
P11	Mean	451.7bc	28.34a	704.0fg	0.34cd	5.95abc
	SD	102.1	21.18	197.7	0.01	0.76
	COV (%)	19.22	51.33	20.16	8.62	35.95
P12	Mean	404.9ab	27.52a	676.3fg	0.29d	8.50cd
	SD	65.2	24.47	184.3	0.02	83.06
	COV (%)	13.45	53.45	27.26	12.65	34.25
P13	Mean	337.7a	26.84a	637.8g	0.19e	9.25d
	SD	55.6	25.15	206.6	00.02	1.77
	COV (%)	13.12	49.44	32.41	13.17	38.73
P_{SL}		*P>0.005*	*P<0.005*	*P<0.005*	*P<0.005*	*P<0.005*

[BT]Board type, [sv]statistical values, [SD]Standard diviation, [COV]Coefficient of variation, [NHS]Nail holding strength, [VSH]Vertical screw holding, [SSS]Surface soundness strength, [JH]Janka hardness [FGD]Formaldehyde gas diffusion, [(*)]According to Duncan's multiple comparison test (P>0.05), there is no statistically significant difference between the mean values indicated by the same letters.

Table 5. Chemical component ratios of the test materials (*Ugur, 2021)*

Test materials	Cellulose (%)	Holocellulose (%)	Lignin (%)
Turkish pine	54.85	78.90	25.46
Cotton stalk	48.38	76.30	19.27
Sunflower stalk	47.52	75.08	19.16
Wheat stalk	51.56	77.89	17.56

While VSH strength values in P11 and P12 group samples exceeded the limit values required in the TS EN 320 (1999), only VSH strength results were able to supply the mentioned limit in P13 group samples. As to in the control samples (group P1), the mechanical measurement results except SSS strength fulfilled the requirements of the mentioned EN 312 (2005). In short, CSR boards and WSR boards, the ratio of technological properties decreased as the percentage of annual plants in the board matrix increased. This result was especially true for 75% added and 100% pure boards. In addition, when the effect of chemical components on the FGR values of test samples is examined on the basis of mixing ratio, there is an opposite effect to that seen on technological properties. Because the lowest FGR values here were measured in SSR samples with the lowest percentage of heolocellulose (75.08%) and the lowest cellulose ratio (47.52%), excluding the 25% cotton stalks FGR value (3.64%).

Although there is a small difference between woods and annual plants (Schafer & Roffael, 2000; Kunaver et al., 2010; Sari et al., 2012; Murata et al., 2013; Lin et al., 2014; Bardak et al., 2019; Peng et al., 2022), in general, lignin enhance FGR values in lignocellulose materials, while holocellulose, especially hemicellulose, has a reducing effect. The average FGR values calculated according to the raw material types from the group's data in Table 4 (WCR: 3.53%, CSR: 6.95%, SSR: 5.55% and WSR: 7.20%) largely comply with the above-mentioned determination.

On the other hand, the course of FGR values (FGR_{WSR}: 7.2% < FGR_{CSR}: 6.95% < FGR_{SSR}: 5.55%), which is the most basic indicator of this article and Table 4, in the total group averages is significantly different according to the echnological properties. As can be understood from this ranking, the best result in terms of FGR was obtained from SSR, and the worst value was obtained from WSR. Figure 2 shows the comparison of the FGR values measured according to the mixture percentages of the test samples obtained from the composite boards produced from WCR-CSR, WCR-SSR and WCR-WSR mixtures with the relevant EN 120 (1996).

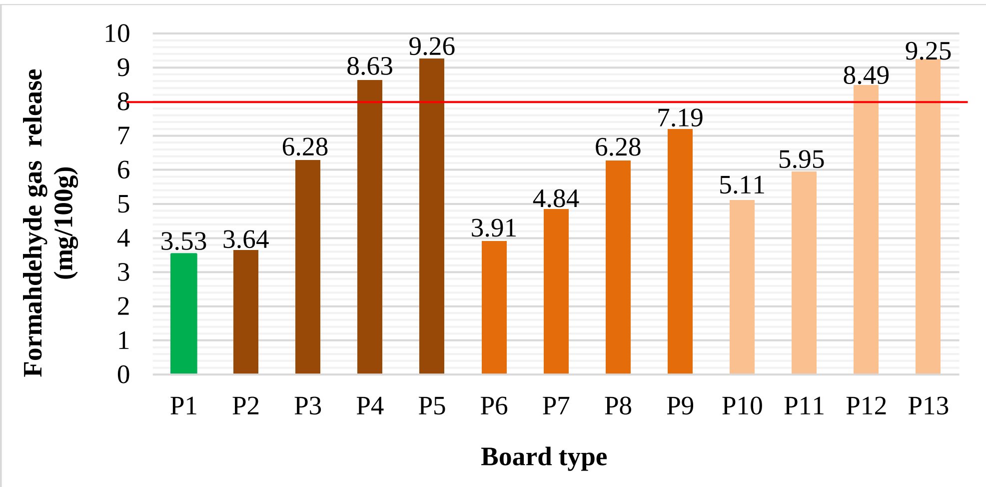

Figure 2. Comparison of formaldehyde gas diffusion percentages with the standard value (8mg $100g^{-1}$)

When the WCR-CSR mixtures in Figure 2 are examined, it will be seen that the FGR values of the P1, P2 and P3 group samples, except for P4 and P5, are below the upper limit value required in EN 120 (1996) (8 mg 100gr^{-1}). The FGR values of the P4 (75%) and P5 (100%) groups samples with higher CSR content were found to be 8.63 and 9.25 (8 mg 100gr^{-1}) higher, respectively, than the other mixtures samples. It can be said that the increase in FGR values of CSR is due to.

In a study on the subject Liang et al. (2021) it was noted that chestnut bark chips effectively reduced FGR values. Nemli and Çolakoglu (2005) stated that mimosa peel particles greatly reduced the formaldehyde emission of particle boards.

As for the WCR-SSR mixtures in Figure 2, the best result in terms of FGR was obtained from these mixture samples. All of the FGR values calculated on the samples belonging to these groups (P6, P7, P8 and P9) were below the upper limit value required in EN 120 (1996). Here, it is a remarkable result that the FGR values (7.19 mg 100gr^{-1}) of the P9 group samples produced from 100% SSR were below the standard value (8 mg 100gr^{-1}). For this reason, in areas of use where environmental sensitivities are prominent, composite materials made from SSR or a mixture can be preferred.

Similarly, Martins et al. (Martins et al., 2020) state that increasing the tannin extract in urea-formaldehyde adhesive reduces free formaldehyde emission in particle boards by 22.5%. As can be seen in Figure 1, results similar to those obtained from WCR-CSR mixture samples were obtained in the FGR analyzes performed for the slab groups consisting of WCR-CSR mixtures. While the FGR values of the P10 and P11 group samples (5.11 and 5.95 mg 100gr^{-1}, respectively) remained below the upper limit value required by the relevant standard, the FGR values of the P12 and P13 groups (8.49 and 9.25 mg 100gr^{-1}, respectively) exceeded the relevant ultimate value.

It can be thought that this negative result for N75 and P00 groups related to FGR value is due to the components of WSR (Table 4) and their known sensitivity to glue. Raw materials with lower FGR values such as WCR, CSR, and tea leaves (Shi et al., 2006) can be added to the mixture in order to reduce the above-standard FGR values emitted from high proportions of WSR mixtures. In a study, Buyuksari et al. (2010) determined that adding cone particles to panels significantly reduced formaldehyde emissions. Besides, Liang et al. (2021) stated that waste chestnut bur can also be used as a natural free formaldehyde cleaner in the production of composite chipboard.

In another study, natural compounds such as Soy protein can react with these binders to reduce formaldehyde release, thus allowing the production of environmentally friendly and human-friendly products. (Perreira et al., 2016).

Conclusions

The water uptake (WU) and thickness increase (TI) measurement results calculated on the test samples showed that the test panels (LIDP) could not meet the criteria required in the standards for use in general purpose and interior designs. In technological properties, also, the strength values decreased as the AWP ratio in the LIDP matrix increased. The best technological strength values in AWP added samples were obtained from 25% CSR added (P2 group) samples, excluding SSS (P9 group 0.68 MPa). Most of the mechanical properties except SSS met the relevant standards requirements.

The FGR values showed "ecofriendly" features, as meeting the requirements of the European Standard, except for the P4 (8.63%), P5 (9.26%), P12 (8.50%) and P13 (9.25%) groups. When the formaldehyde gas releases measured in the experiments were evaluated according to European Standard, the best results were obtained in the samples manufactured from WCR-SSR mixtures. The most striking result in terms of gas emissions in the LIDP panels produced is the FGR value (7.19 mg 100gr^{-1}), which complies with the standard limits measured in samples obtained from pure sunflower stalks (P9 group).

Based on the test results, if the physical and technological properties of the LIDP panels obtained, as well as their FGR values, meet the required conditions in the relevant standards, they can be recommended for use in areas where environmental sensitivities come to the fore.

Ethics Committee Approval

N/A

Peer-review

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Author Contributions

Conceptualization: C.U., İ.B.; Investigation: C.U., İ.B. ; Material and Methodology: C.U.; Visualization: İ.B.; Writing-Original Draft: C.U.; Writing-review & Editing: C.U., İ.B.; The author has accepted to publish the version of manuscript.

Conflict of Interest

The authors declare that they have no conflict of interest.

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