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### Research Article

## UTILIZATION OF UREA POWDERS WITH DIFFERENT SIZES AS A FORMALDEHYDE-SCAVENGER IN THE PARTICLEBOARD MANUFACTURING

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### ABSTRACT

In this study, the effects of different size and rate of urea powder usage in particleboard manufacturing on the formaldehyde emission of the boards were investigated. Two different sizes (Large, Small) and five different rates of urea powder (1, 2, 3, 4 and 5%) were used for particleboard manufacturing. Urea formaldehyde (1.35 moles) adhesive was used for production of three layered particleboards. Formaldehyde contents were determined by perforator method according to EN 120. Furthermore, mechanical and physical properties including bending strength, modulus of elasticity, internal bond strength, surface stability, thickness swelling and water absorption of the samples were determined according to EN 310, EN 319 and EN 317 standards, respectively. Formaldehyde emission values were decreased with the mixing of the urea powder with chips prior to gluing and the produced boards had E0 grade in terms of formaldehyde emission. The size and rate of the urea powder were statistically effective on the mechanical and physical properties of the produced boards. In addition, all of the boards produced with small size urea powders satisfied the required standards for mechanical, physical and formaldehyde emission properties, except groups produced with 5% small size urea powder. It should be noted that slight decrease of mechanical and physical properties were observed with the loading of urea powder. As a result, it was determined that using of the small size urea powder provided better results than large size.

**Keywords:** Formaldehyde emission, carcinogenic substance, perforator method, particleboard, urea formaldehyde adhesive, urea powder.

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## 1. INTRODUCTION

Due to the rapid growth of the world population, demand in the furniture industry has increased. With this increase, the need for raw materials has also raised. Particleboard and MDF are the most used wood-based boards in the furniture industry. Turkey has an important place in Europe and the World for the wood-based panel sector and this is a fast-developing sector in Turkey. Formaldehyde-based adhesives such as urea-formaldehyde (UF) and melamine-formaldehyde (MF) resins are the most commonly used adhesives in the manufacturing of these wood-based boards. Using the formaldehyde-based resins causes some disadvantages for these kinds of boards. The main and also the most important disadvantage of wood panels produced with formaldehyde-based resins is their formaldehyde emission which is identified as “probably carcinogenic to humans” (Group 2A) by International Agency for Research on Cancer (IARC) in 1995 (IARC 1995) [1]. In addition, the definition was changed from Group 2A- “probably carcinogenic to humans” to Group 1- “carcinogenic to humans”-“formaldehyde is carcinogenic to humans” by IARC (IARC 2006) [2]. One of the main issues of panel manufacturers is solving this problem. Researchers have been performed many studies on this area.

The most common method is the use of resin with a lower mole ratio for reducing formaldehyde emission values. However, using lower mole ratio adhesive decreases the formaldehyde emission values at the expense of strength values of the particleboards. Usage of hardeners, fillers, and additives prepared in proper formulations in order to scavenge free formaldehyde after hardening helps to improve formaldehyde emission properties [3]. Some researchers aimed to reduce the formaldehyde emission values by using tannins in urea-formaldehyde resin [4,5]. The use of tannins in urea formaldehyde resin has helped to reduce the formaldehyde emission values of the particleboards. Besides, the addition of melamine in the second or third stage of urea-formaldehyde adhesive manufacturing is another way to decline formaldehyde emission. The reduction of free formaldehyde in the panels with melamine powder is a known fact, but the rising of melamine powder usage caused to increasing of resin cost [6]. In addition, one of the other methods is using formaldehyde-scavenger chemicals in the mat during the manufacturing of the particleboards or the resin for the decrease formaldehyde emission values. Chemicals most commonly used as formaldehyde-scavengers are amine-based ones. Boran et al. (2011) [7] also added amine compounds in UF resin and produced medium-density particleboards. It was determined that formaldehyde emission from medium-density particleboard panels decreased by adding urea, propyl amine, methylamine, ethylamine, and cyclopentyl amine solution. Furthermore, Atar et al. (2014) [8] used a water solution of urea powder (10 wt%) as a formaldehyde scavenger in UF resin. It was reported that usage of 1 wt% of the solution based on the solid weight of the UF resin decreased the formaldehyde emission values but slight decrease in mechanical and physical properties was also observed. In the literature, studies on the comprehensive usage of urea powder as a formaldehyde-scavenger are quite shallow, except in resin manufacturing.

In this study, utilization of urea powders with different sizes and different rates as a formaldehyde-scavenger in the particleboard manufacturing was investigated. For this purpose, three-layer particleboards were manufactured with two different sizes (Large, Small) and five different rates of urea powder (1, 2, 3, 4, and 5%). Formaldehyde content (by Perforator method), mechanical and physical properties of the samples were determined according to EN 120, TS EN 310, TS EN 319, TS EN 311, and TS EN 317 standards, respectively.

## 2. MATERIALS AND METHODS

Urea formaldehyde (UF) resin with a molar ratio of 1.35 (with 62% solid content) was used as an adhesive for manufacturing of three-layer particleboards. Coarse and fine chips consisting of a mixture of red pine and poplar wood supplied from Kastamonu Integrated particleboard facilities

(Tarsus/Turkey) were used. Commercial urea obtained from Comzest Trading Fzc, which is used in manufacturing of adhesive for particleboard and MDF production, was used as urea powder. Ammonium chloride supplied from Akça Chemical Substances, Transportation, Trade Industry Incorporated Company was used as a hardener for UF resin. Aqueous solution of Ammonium chloride (with 25% solid content) was prepared as a hardener.

## 2.1. Classification of Urea Powder

Urea powders were screened with automatic vibrating sieve machine and passed from 0.2 mm sieve was used as small size urea powder. Urea powders in sizes between 1mm and 0.2mm sieve were used as large sized urea powder. (Included stayed on 1mm and 0.2mm sieve).

## 2.2. Manufacturing of Particleboards

Fine particles were utilized in surface layers (SL) while coarse ones in core layer (CL). Eleven different particleboard groups with three layers (two surface layers and one core layer) were manufactured. The experimental design of the study was presented in Table 1. The core layer was accounted for 67% of the total board weight. Surface layers were contained 33% of the total board weight.

**Table 1.** Experimental Design

ID	Urea Formaldehyde Resin (%) <sup>*</sup>	Urea Powder Size	Urea Powder Amount (%) <sup>**</sup>	Amonium Chloride (%) <sup>***</sup>
Control	10	-	-	10
S1	10	Small	1	10
S2	10	Small	2	10
S3	10	Small	3	10
S4	10	Small	4	10
S5	10	Small	5	10
L1	10	Large	1	10
L2	10	Large	2	10
L3	10	Large	3	10
L4	10	Large	4	10
L5	10	Large	5	10

\*Same rate of Resin was used in both layers.

\*\*Based on Dry Resin amount

\*\*\*Ammonium Chloride with 25% solids content was used based on the liquid amount of adhesive.

Depending on the Experimental design given from Table 1, first particles and urea powders were dry-mixed in a high-intensity mixer. Then, UF resin which has hardener added into the high-intensity mixer to produce a homogeneous blend. The blends were laid into frame of 500mm x 500mm. A hot press was used for forming of particleboards (90-120 Bar). The target thickness was 19mm. Pressing time and temperature were 210s and 205 °C, respectively. After pressing, particleboards were conditioned at a temperature of 20 °C and 65% relative humidity. The

conditioned boards were cut from four edges and grinded thickness range of 0.50 – 1.00 mm. Then test samples were cut according to TS EN standards.

### 2.3. Testing of Manufactured Boards

Testing of the samples was conducted in a climate-controlled testing laboratory. Densities were measured by air-dried density method according to the TS EN 323 standard. Bending strength, modulus of elasticity, internal bond strength, surface soundness, screw withdrawal strength, thickness swelling and water absorption of the samples were determined according to TS EN 310, TS EN 319, TS EN 311 and TS EN 317 standards, respectively. Mechanical properties testing were performed on Zwick Z010 (10KN).

### 2.4. Analysis of Data

Design-Expert® Version 7.0.3 statistical software program was used for statistical analysis. The effectiveness of urea powder rate and size as a formaldehyde-scavenger in particleboard manufacturing was evaluated.

## 3. RESULTS AND DISCUSSION

Moisture Content after the Pressing (Table 2.) and Means of Density (Table 3.) were given in the tables below.

**Table 2.** Moisture Content after the Pressing

Grup	Number of Samples	Mean of Moisture Content (%)
Control Board	3	<b>6,20</b>
Small	18	<b>6,00</b>
Large	18	<b>6,52</b>

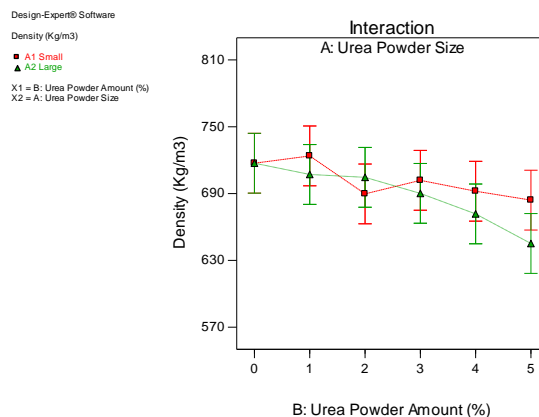
The average moisture content of the produced particleboards after pressing varies between 6.00% and 6.52%. The highest average moisture content (6.52%) was observed from groups where the large size urea powders were used. In addition to that, the lowest average moisture content (6.00%) was observed from groups which contained small size urea powders. However, small size contained groups shows closed average moisture content with control groups (6.20%).

**Table 3.** Means of Density

ID	Number of Samples	Mean Density (kg/m <sup>3</sup> )
Control Board	8	<b>687,60</b>
Small	40	<b>697,85</b>
Large	40	<b>679,26</b>

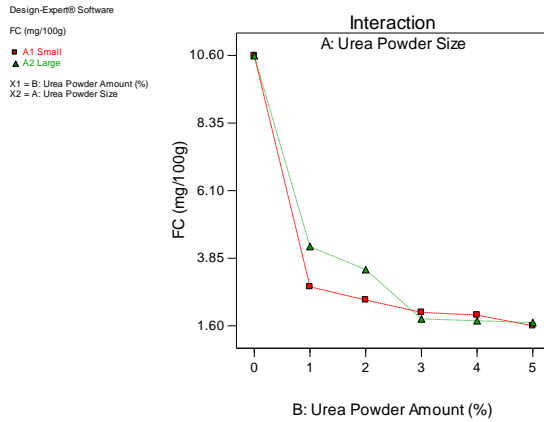
When the Table 3 examined, it is observed that the average density values of the produced particleboards were close to each other. The close results to the targeted board density values were obtained. Interaction graph of Density values was also shown in Figure 1. If we handled Figure 1, it is seen that board densities were slightly decreasing with the amount of urea powder increases. However, the effect of urea powder size (P = 0.2728) and urea powder amount (P = 0.0625) on this change was not found to be statistically significant.

As can see from the interaction graph given in Figure 2, when the first adding of urea powder Formaldehyde Content (FC) was sharply declined. Decreasing on FCs were continued with the loading of urea powder. However, after first loading, every 1% added was not as effective on the FC as the first addition. The maximum allowable formaldehyde content for E0, E1, and E2 class particleboard is 2, 8, 30 mg/100g dry particleboard sample according to EN 312, respectively. The control group boards (10.59 mg/100g) in the E2 Formaldehyde emission class were upgraded to E0 class by using 5% small urea powder (1.6 mg/100g). Costa et al. (2013) used urea powders as a formaldehyde scavenger (FS) with three different rates (5%, 10%, and 15%) in the particleboard manufacturing in their study [9]. The formaldehyde content of the particleboards produced with low mole ratio Urea Formaldehyde resin decreased with urea powder using. Urea powders have been successfully evaluated as a FS. However, it has been stated that urea powders used without any dimensional classification were not as effective as other FSs used. Atar et al. also used 1% urea powder without dimensional classification as a FS in the production of particleboard. In that study, it was noted that urea powder was effective in decreasing the formaldehyde content and lower formaldehyde content was obtained compared to the control samples [8]. Boran et al. (2011) were also used urea as a FS in the manufacturing of MDF panels [10]. Urea was successfully utilized as a FS and formaldehyde emission values were diminished. Large urea powder groups showed parallel results with small one. It has been determined that the size of the urea powder has a slight effect on formaldehyde emission values.

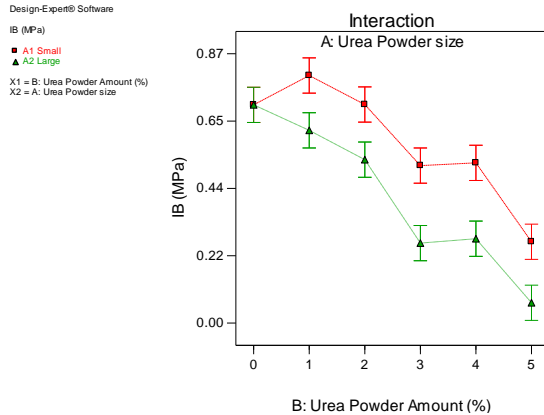


**Figure 1.** Interaction graphs of Density.

Based on results, both urea powder size and amount had significant effect on IB strength values ( $P < 0,0001$ ). Interaction graphs of internal bond strength were shown in Figure 3. With the adding of urea powder IB values were reduced. Costa et al. (2013) and Atar et al. (2014) were also reported that IB values were declined with using of urea powder [8,9]. In addition, it has been stated that IB values decreased with the usage of urea as a FC in MDF production [10]. Furthermore, IB values were declined with usage of different FS in the manufacturing of different type wood-based panels [11,12]. Small size urea powder group were provided better IB properties than the large ones. The all board produced with small size urea powder groups satisfied standard requirements (P2 class particleboard: furniture boards for the interior application) for IB properties (0.35 MPa), except 5%. For the 1% and 2% large size groups satisfied the standards but others groups not. In the groups where 3% and more large size urea powder was used, much lower IB values observed compared to those using 1% and 2%.



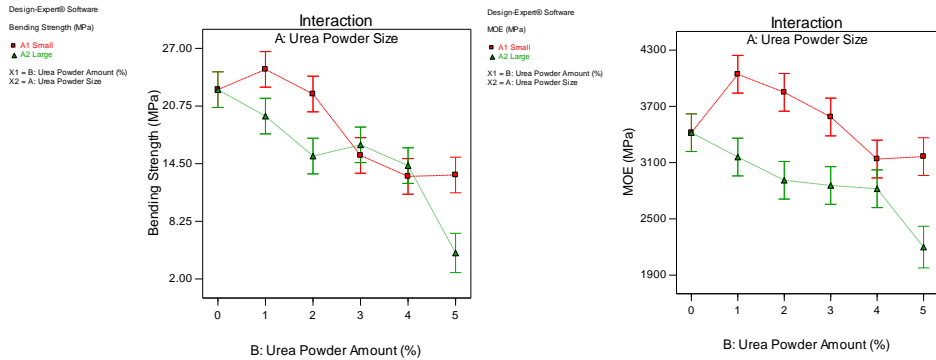
**Figure 2.** The influence of urea powder size and rate on the formaldehyde content.



**Figure 3.** Interaction graphs of internal bond strength.

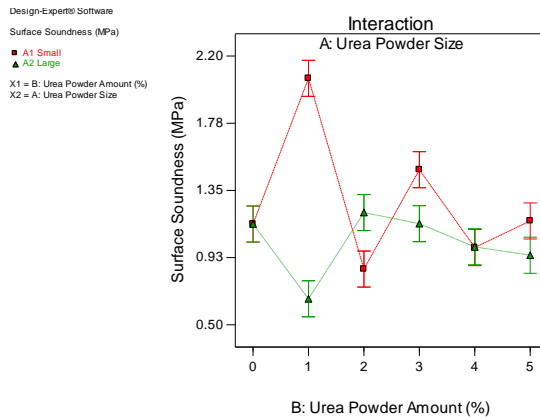
The interaction graphs of bending strength and modulus of elasticity were given in Figure 4. It was determined that the urea powder size had a statistically significant effect on the bending strength ( $P = 0.0013$ ). Larger urea powder sizes decreased the bending strength values. Better results were obtained from boards produced using small size urea powder than large size urea powder boards. It was observed that the amount of urea powder has also statistically affected the bending strength values ( $P < 0.0001$ ). A decrease in bending strength was observed with the increase in the amount of urea powder. But all the manufactured boards were provided standard requirements (11 MPa) for P2 class boards in the standard, except large size with 5% using.

In the MOE properties, parallel results were observed with bending strength properties. As the size and amount of urea increased, the MOE tended to decline. Besides the results, the size and amount of urea were significantly effective on the MOE ( $P < 0.0001$ ). As with bending strength, all the produced boards were provided standard requirements (1600 MPa) for P2 class boards in the standard, except large size with 5% using. In addition, in that group, a modulus of elasticity (1567.99 MPa) close to the standard was obtained. For the bending properties similar results were reported in the literatures [8,10].



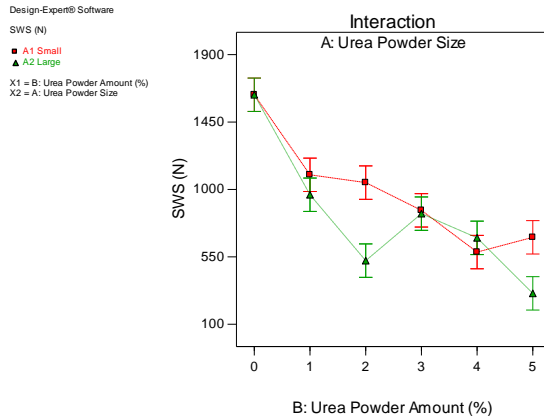
**Figure 4.** The effects of urea powder size and rate on the bending properties.

Size of the urea powder has a statistically significant effect on the surface strength feature ( $P < 0.0001$ ). Small size Urea powder groups provided better surface strength values than large sizes. The amount of urea powder has a significant effect on surface strength values ( $P = 0.0002$ ). All the produced board groups showed higher results than the standard value ( $\geq 0.8$  MPa) required for P2 class particleboards.



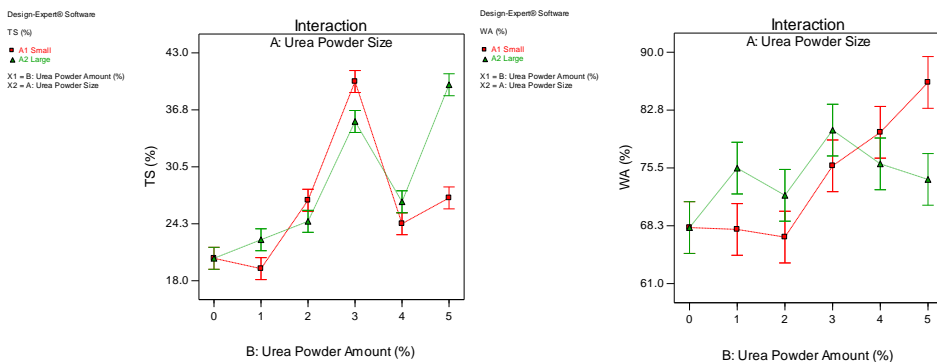
**Figure 5.** Interaction graphs of the surface soundness.

The maximum force was determined in the SWS test. Similar to the MOE values, all the produced boards reached the standard requirements for SWS, except for samples having 5% large size urea (min. 450 MPa). Urea powder size ( $P=0.0014$ ) and amount ( $P<0.0001$ ) had significant effect on SWS properties. Better results were obtained in groups where small size urea powder was used compared to the large size urea powder ones. SWS values were decreased with the increase of the urea powder amount.



**Figure 6.** Interaction graphs of screw withdrawal strength.

Thickness swelling (TS) and water absorption (WA) tests were carried out as physical properties of the produced boards. As a result of the test, the interaction graphs of TS and WA were given in Figure 7. Amount ( $P < 0,0001$ ) and size ( $P = 0,0006$ ) of the urea powder were significantly effective on TS properties. TS properties were getting worst with the rising of urea powder amount. As it is mentioned previously, with the increase of the urea powder amount, the quality of adhesion in the core layer was reduced leading to lower IB values. Lack of good adhesion may be facilitated the water penetration into the boards during the TS test and may cause an increase in thickness swelling values. The water absorption interaction graph was also presented in Figure 7. Some changes were observed in the WA values for both groups. It was determined that urea powder size had no statistical effect on these changes ( $P = 0.8682$ ), but the amount of urea powder had a statistically significant effect on the WA feature ( $P < 0.0001$ ). While none of groups were satisfied the standard for TS (Max. 15%), all groups provided standard requirements for WA (Max. 80%), except small size urea powder with 5% (86.21%). Similar results were observed for TS [9] and WA [10] in the previous study.



**Figure 7.** Interaction graphs of thickness swelling and water absorption (at 24h).

Moreover, all the data and standard requirement were summarized in Table 4. While values matched standards were painted in green, not matched values were painted in red.



**Table 4.** Summary of Study

ID	FC	IB	BS	MOE	SS	SWS	WA	TS
<b>Control</b>	10,59 E2	0,70 (0,06)	13,34 (0,32)	3418,96 (460,09)	1,14 (0,03)	1064,67 (177,94)	68,02 (6,02)	20,46 (1,26)
<b>S1</b>	2,90 CARB2	0,80 (0,06)	14,01 (0,12)	4042,04 (141,96)	1,12 (0,14)	1097,67 (130,70)	67,79 (2,79)	19,33 (0,88)
<b>S2</b>	2,46 CARB2	0,71 (0,10)	13,82 (0,78)	3849,33 (353,78)	0,85 (0,03)	1045,33 (290,29)	66,83 (4,16)	26,83 (1,73)
<b>S3</b>	2,05 CARB2	0,51 (0,10)	13,11 (0,04)	3586,31 (32,70)	1,48 (0,21)	860,00 (94,50)	75,76 (4,07)	39,86 (82,32)
<b>S4</b>	1,96 E0	0,52 (0,10)	13,29 (0,27)	3137,68 (158,47)	0,99 (0,25)	581,67 (15,57)	79,96 (5,12)	24,26 (1,26)
<b>S5</b>	1,60 E0	0,26 (0,09)	13,58 (0,11)	3163,91 (40,04)	1,16 (0,20)	680,00 (55,07)	86,21 (5,31)	27,09 (1,23)
<b>L1</b>	4,24 E1	0,62 (0,08)	13,89 (0,09)	3159,23 (15,18)	1,20 (0,06)	964,33 (171,10)	75,47 (5,32)	22,50 (1,13)
<b>L2</b>	3,47 CARB2	0,53 (0,06)	13,23 (0,01)	2911,46 (35,50)	1,21 (0,15)	524,67 (4,51)	72,06 (0,70)	24,52 (1,18)
<b>L3</b>	1,83 E0	0,26 (0,01)	13,92 (0,09)	2855,90 (11,00)	1,14 (0,05)	838,33 (25,70)	80,24 (1,49)	35,47 (1,37)
<b>L4</b>	1,77 E0	0,27 (0,03)	13,38 (0,27)	2821,21 (197,12)	0,99 (0,08)	676,67 (56,39)	76,01 (1,62)	26,66 (0,60)
<b>L5</b>	6,19 E1	0,07 (0,00)	13,31 (0,20)	1567,99 (115)	0,94 (0,11)	306,00 (8,54)	74,06 (1,34)	39,50 (2,09)
<b>Standard Value</b>	E1 ≤ 8.00*	≥ 0.35	≥ 11	Min. 1600	≥ 0.80	≥ 450	Max. 80	Max. 15

\*According to the EN 120 perforator method which stays in EN 13986 standard for European Countries, E1 limit for wood based boards such as particleboard and MDF.

From Table 4, it was clearly seen that the size and amount of urea powder usage have a significant effect on the board's properties. About the formaldehyde content, E0 and E1 class particleboard produced with different size urea powders. All the produced groups satisfied the E1 class particleboards standard, except for control group. In addition, some groups having small size (S4 and S5) and large size (L3 and L4) urea powders provided E0 class particleboards. All boards produced with small size urea powder were satisfied standard requirements for all mechanical properties, except for S5 group. The boards produced with large size urea powder up to 4% were fulfilled the standard requirements for mechanical properties, except IB properties. L1 and L2 groups provided much higher IB values than the standard values. These two groups were also satisfied the all other mechanical properties required by standards. To mention on physical properties, while none of groups were satisfied the standard for TS, all groups provided standard requirements for WA, except small size urea powder with 5%. L3 group boards showed the maximum WA value of 80%. It should be noted that there was no paraffin or equivalent products was used in this study. These physical property results might be improved by appropriate addition of water repellent chemicals.

#### 4. CONCLUSION

As results of the study, different size and amount of urea powder were successfully utilized as a formaldehyde-scavenger in the manufacturing of particleboards with UF resin and the following conclusions were reached;

1. The best results were obtained by using 5% of small size urea powders for Formaldehyde content,
2. With the presence of a small amount of urea powder in the formulation, Formaldehyde emission was sharply declined. Further, the increase in urea powder amount provided moderate improvement not as effective as the first addition,

3. Both the amount and the size of urea powders had significant effect on panel properties,
4. The physical properties of the some of the produced samples were not satisfied the standard requirements. Using of some water repellent chemicals might help to overcome that problem.

As a result of the studies, it has been observed that urea powder can be used as a formaldehyde-scavenger. Reducing the formaldehyde emission, which is dangerous for human health, is one of the main goals of every manufacturer. It is thought that this study can be guide for wood-based board manufacturer. It should be investigated whether the studies are suitable for mass production by working more and cost studies should be done.

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### REFERENCES

- [1] International Agency for Research on Cancer (IARC) (1995) Wood dust and formaldehyde. IARC Monogr Eval Carcinog Risks Hum, 62:1–405.
- [2] International Agency for Research on Cancer (IARC) (2006) Formaldehyde, 2-butoxyethanol and 1-tertbutoxypropan-2-ol. IARC Monogr Eval Carcinog Risks Hum, 88:1–478. PMID:17366697.
- [3] Şahin H.T., Filiz M., Kaya A.İ., Usta P., Çiçekler M., Bozkurt C. (2011) Formaldehid Tutkalı ve Formaldehid Emisyonu. Laminart, Sayı:39.
- [4] Beer J., Schmidt H. and Bistram E.V. 1994. Method for the Fabrication of Particle Boards and Fibreboards with Low Formaldehyde Content Using Tannin Based Binders. European Patent Office. Patent number: EP0788866B1
- [5] Çolak S., Öztürk H., Demir A., 2009. Yonga Levhaların Formaldehit Emisyonu Üzerine Tanen ve Kitosanın Etkileri.
- [6] Pizzi A. 1994. Advanced Wood Adhesives Technology. Marcel Dekker, Inc.
- [7] Boran S., Usta M., Gümüşkaya E. 2011. Decreasing Formaldehyde Emission from Medium Density Fiberboard Panels Produced by Adding Different Amine Compounds to Urea Formaldehyde Resin. International Journal of Adhesion and Adhesives, 31(7):674-678.
- [8] Atar İ., Nemli G., Ayrılmış N., Baharoğlu M., Sarı B. and Bardak S. 2014. Effects of Hardener Type, Urea Usage and Conditioning Period On the Quality Properties of Particleboard. Materials & Design (1980-2015) 56, 91-96.
- [9] Costa N.A., Pereira J., Ferra J., Cruz P., Martins J., Magalhaes F.D., Mendes A. and Carvalho L.H. 2013. Scavengers for Achieving Zero Formaldehyde Emission of Wood-Based Panels. Wood Sci Technol (2013) 47:1261–1272.
- [10] Boran, S., Usta, M. and Gümüşkaya, E. 2011. Decreasing formaldehyde emission from medium density fibreboard panels produced by adding different amine compounds to urea formaldehyde resin. International Journal of Adhesion and Adhesives 31: p. 674-678.
- [11] Boran, S., Usta, M., Ondaral, S. and Gümüşkaya, E. 2012. The Efficiency of tannin as a formaldehyde scavenger chemical in medium density fibreboard. Composites: Part B 43: p. 2487-2491.
- [12] Ghani, A., Bawon, P., Ashaari, Z., Wahab, M.W. and Hua, L.S. 2017. Addition of Propylamine as Formaldehyde Scavenger for Urea Formaldehyde-Bonded Particleboard. WOOD RESEARCH 62 (2): p. 329-334.