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THE USAGE OF ABCM AS A FORMALDEHYDE SCAVENGER DURING PARTICLEBOARD MANUFACTURING WITH MELAMINE IMPREGNATED PAPER WASTE (MIPW)

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Abstract:
Particleboards are extensively used in indoor applications due to their some advantages over massive wood. Particleboards, produced with formaldehyde-based adhesives or that kind of adhesives wastes (Urea-formaldehyde, Melamine-formaldehyde, melamine impregnated paper waste etc.), had high level formaldehyde emissions. In our recent study, particleboards, manufactured with MIP wastes as a resin replacement, showed much higher formaldehyde emission values than standard requirements and that motivated to perform this study. In this study, Amine based chemical mixture (ABCM) was used as a formaldehyde scavenger during particleboard manufacturing. Melamine impregnated paper wastes (MIPW) were used as adhesive. The effect of ABCM amounts on the formaldehyde content and other panel properties were investigated. Based on our previous findings, MIPW 25% was used for both core and surface layers. Five different rates of ABCM (0, 2.5, 5, 7.5 and 10%) were studied. Formaldehyde content was examined by perforator method according to EN 120. Furthermore, mechanical and physical properties including 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 EN 310, EN 319, EN 311, EN 320 and EN 317 standards, respectively. The experimentally results showed that, amount of ABCM had significant effect on formaldehyde content. With the increasing of ABCM rates, formaldehyde values was decreased. The usage of 10% ABCM provided the best results. All the tested boards had satisfied the standard requirements for mechanical and physical properties except thickness swelling and formaldehyde content.

Key words: *formaldehyde emission; amine based chemical mixture; perforator method; melamine impregnated paper waste; mechanical and physical properties.*

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INTRODUCTION

Demands for wood and wood products are increasing day by day due to the rapid growth of the world population. This increase needs more wood materials. As one of the most common problems seen in today's forest destructions may be able to avoided by means of the rational use of forests and forest product (Başboğa and Güntekin 2016). The alternative wood products such as particleboards, medium density fiber board (MDF), oriented strength board (OSB) etc. were produced instead of solid wood for sustainable use of forests. Lignocellulosic materials such as firewood, sawdust, annual plants stalks, forest pruning wastes are used as well as wood raw materials in the particleboard manufacturing. Furthermore, formaldehyde based resins such as urea-formaldehyde, melamine-formaldehyde, and phenol- formaldehyde resins were used as an adhesive. 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). 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). Solving this problem is one of the main issues of panel manufacturers. Many studies have been carried out on this topic.

The addition of some formaldehyde-scavenger chemicals to the mat during the manufacturing of the particleboards with urea-formaldehyde resin reduces the formaldehyde emission. The addition of extra urea, phenol or melamine powder during resin production allows free formaldehyde to be trapped. The most common and simple method is to apply the formaldehyde / urea ratio closer to 1 or 1 in the mixture of resin, but this process affects negatively to panels strength values. Because of that the press time must be kept longer than standard. In addition, long press time causes to capacity loss in production (Lee and Chung 1984). The addition of melamine in the second or third stage of urea-formaldehyde resin manufacturing is another way to reduce formaldehyde emission. The reduction of free formaldehyde in the panels with melamine powder is a known fact, but the increasing of melamine powder usage caused to rising of resin cost (Pizzi 1994). Usage of hardeners, fillers and additives prepared in proper formulations in order to scavenged free formaldehyde after hardening helps to improving formaldehyde emission properties (Şahin et al. 2011).

In this study, the usage of amine based chemical mixture (ABCM) as a formaldehyde emission scavenger during particleboard manufacturing with melamine impregnated paper waste (MIPW) as resin was investigated. On this purpose, three-layer particleboards were manufactured from wood particles mixture adding MIPW and ABCM. 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.

MATERIAL AND METHODS

Materials

Melamine impregnated paper wastes (MIPW) as adhesive and two different types of particles (fine and coarse) were used. Turkish red pine (90%) and poplar wood (10%) particle mixtures were used. Amine based chemical mixture (ABCM) was used as a formaldehyde scavenger. MIP waste was obtained from the impregnation line of Kastamonu Integrated Adana MDF Facility and ABCM was prepared in the laboratory of the same facility.

Particleboard Manufacturing

Melamine impregnated paper wastes (MIPW) were granulated in Pulverizator with cooling capabilities, into the flour form. These flours, screened and passed from 0.2mm sieve, were used as an adhesive in this study. Fine particles were utilized in surface layers (SL) while the coarse ones in the core layer (CL). Five different groups were made. Three particleboards with three layers (two surface layers and one core layer) were manufactured for each group. The experimental design of the study was presented Table 1. The core layer was accounted for 67% of the total board weight. Surface layers were contained 33% of the total board weight. Based on our previous findings, MIPW 25% was used for both core and surface layers (Başboğa et al. 2016).

Table 1

Manufacturing schedule of Particleboards

Board-Type	MIPW (%)	ABCM (%)*
A0	25	0.0
A2.5	25	2.5
A5	25	5.0
A7.5	25	7.5
A10	25	10.0

* According to the ratio of liquid resin

MIPW used in this study had 52% resin containing approximately 50% Urea-formaldehyde (UF) and 50% Melamine-formaldehyde (MF) solid. The given rate of MIPW in the Table 1 was estimated as a liquid resin with 65% solid content. The amount of ABCM was determined by liquid weight ratio of MIPW. Depending on the formulation, the wood particles, MIPW and ABCM were dry-mixed in a high-intensity mixer to produce a homogeneous blend. Same ratio of MIPW and ABCM were used in both core and surface layers. The blends were laid into a frame of 500mm x 500mm. An automatic hot press with computer controlled was used for forming the particleboards. The target thickness was 19mm. "Reach the target thickness in forty seconds, "then keep thickness from target" was set as a computer command. Maximum applied pressures were reached in forty second then it was automatically decreased. The applied pressures were at 0.4-6.2MPa range. Pressing time and temperature were 240s and 200°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 sanded until their thickness was 18mm. Then test samples were cut according to TS EN standards.

Particleboard testing

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. Formaldehyde content was examined by perforator method according to EN 120. Furthermore, mechanical and physical properties including 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 EN 310, EN 319, EN 311, EN 320 and EN 317 standards, respectively. Three boards for each group were made and three samples were tested for each board to determine each property. Nine samples were examined for all properties testing in total. Mechanical properties testing were performed on Zwick Z010 (10KN).

Statistical analysis

Design-Expert® Version 7.0.3 statistical software program was used for statistical analysis. ANOVA test was used to determine the effects of the factors. The effectiveness of ABCM as a formaldehyde scavenger in particleboard manufacturing was evaluated.

RESULTS AND DISCUSSION

Particleboards were produced in the density range of 755-798kg/m^3. In this study, formaldehyde content, mechanical (bending strength, modulus of elasticity, internal bond strength, surface soundness and screw withdrawal strength) and physical (thickness swelling and water absorption) properties of all samples were determined. All data of the study were summarized in Table 2. The average value and standard deviation values were given for each group.

Table 2

Summarize of mechanical, physical and formaldehyde content

ID	Formaldehyde content (mg/100g oven dry board)	Bending strength (MPa)	Modulus of elasticity (MPa)	Internal bond strength (MPa)	Surface soundness (MPa)	S.W.S* Average maximum load (N)	Thickness swelling (at 24h) (%)	Water absorption (at 24h) (%)	Moisture content (%)
A0	39.88 (E3)	28.45 (1.24)**	4110.08 (116.00)	0.66 (0.08)	0.95 (0.12)	1108.33 (106.01)	17.28 (0.84)	64.93 (7.21)	6.86 (0.08)
A2.5	19,80 (E2)	19.39 (2.11)	3098.17 (299.18)	0.60 (0.05)	1.42 (0.10)	911.00 (130.91)	16.05 (1.89)	62.40 (3.78)	6.04 (0.24)
A5	16.70 (E2)	22.66 (2.21)	3371.56 (72.30)	0.64 (0.07)	1.48 (0.10)	857.33 (74.77)	17.27 (2.35)	62.40 (5.99)	5.85 (0.16)
A7.5	15.13 (E2)	16.90 (0.20)	3016.94 (77.64)	0.57 (0.14)	1.59 (0.47)	734.33 (266.60)	14.39 (1.21)	61.39 (3.60)	6.66 (0.17)
A10	9.05 (E2)	23.27 (2.41)	3697.79 (74.27)	0.54 (0.25)	1.73 (0.16)	669.00 (255.09)	18,55 (5.36)	64.86 (5.26)	5.91 (0.09)
Std	E1 ≤ 8.00***	≥ 11	Min. 1600	≥ 0.35	≥ 0.8	≥ 450	Max. 15	Max. 80	

* Screw withdrawal strength properties
** Values in parenthesis are standard deviations.
***According to the EN 120 perforator method which stay in EN 13986 standard for European Countries, E1 emission limit for wood based boards such as particleboard and MDF.
Std: Standard values

The graph of formaldehyde content (FC) was given in Fig. 1. According to graph, formaldehyde emission values were decreased with addition of amine based chemical mixture (ABCM). E1 limit for wood based boards such as particleboard and MDF is set to 8mg/100g oven dry board as specified in EN 13986 for European countries, based on Perforator test. FC average value which observed from A0 boards (produced without ABCM) was 39.88mg/100g oven dry board. This result was nearly five times bigger than the standard value. However, 9.05mg/100g oven dry board FC average value was observed from A10 boards which produced with highest ABCM rate. That value was in E2 emission limits range and it was closer to E1 release class. In the reaction between formaldehyde and urea, formaldehyde is linked to the amine groups contained in the urea to give (-mono, -di, -tri methylol urea) different numbers of derivatives which has methylol groups (Anonymous 1). It is thought that amine groups in ABCM were linked with free formaldehyde which came out after press from MIPW.

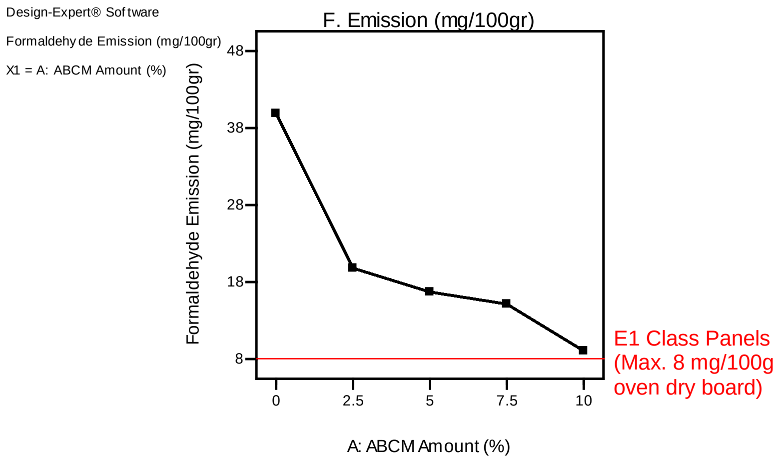

Fig. 1.
The influence of ABCM rate on the formaldehyde content.

The Interrelation of ABCM usage rate between density values was given in Fig. 2. Statistical analysis showed that ABCM amount was not significantly effective on density (P=0.1734). Density was slightly decreased with first loading of ABCM (2.5%) as can be seen in Fig. 2. In contrast with first loading rate, the others were not significantly changed the density values. Densities of all produced panels were determined at the same range, while control groups were slight higher than others.

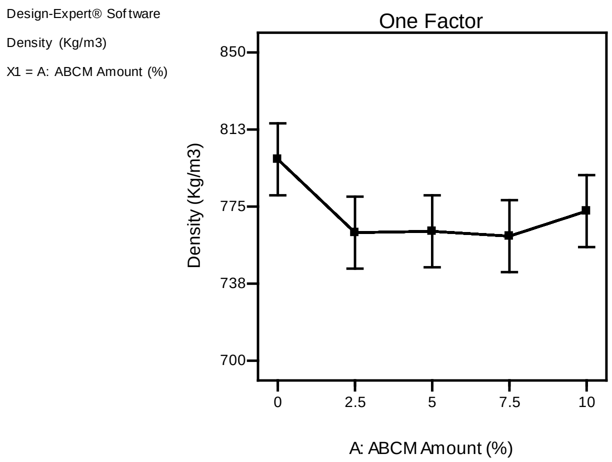

Fig. 2.
Interaction graphs of density.

Effect of ABCM on internal bond (IB) strength was given in Fig. 3. With the first loading amount of ABCM, IB strength value was slightly affected and slight decreasing was observed. With rising of ABCM amount, significant change was not observed on IB strength values. IB values of all groups produced with ABCM were determined at the same range. Although ABCM loading had no significant effect on IB strength (P=0.8100), these strength values were tend to fall. The images of the IB test are shown in Fig. 4. The highest IB strength result was obtained from A0 boards (0.66MPa). However, all produced boards were provided standard requirements (0.35MPa). A10 boards had the lowest IB value (0.54MPa) at the highest percentage of ABCM (10%) and pass the standard limit.

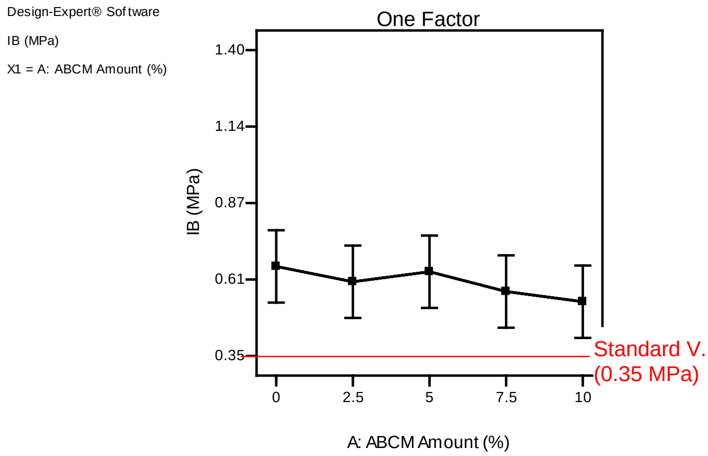

Fig. 3.
Interaction graphs of internal bond strength.

Fig. 4.
Internal bond test.

Changing on screw withdrawal strength (average maximum load) (SWS) values with ABCM loading was shown in Fig. 5. The SWS test (parallel to the surface) is applied from core layer, which has the lowest density and therefore the weakest part of the particleboard. In this layer, particle size is big, adhesive ratio, adhesion and density are lower (Örs et al. 2004). A better adhesion in the core layer will increase the density of this layer and also improve the SWS values. SWS (parallel to the surface) values show parallel results with IB strength. Özen and Efe reported in 1993 that SWS is directly proportional to the density. Therefore, when the IB strength was examined (Fig. 3), it is seen that they show parallel results with the SWS. Amount of ABCM was not statistically effective on SWS properties (P=0.1033) and SWS was shown a tendency to decline. SWS properties were slightly decreased with the rising of ABCM. On the other hand, all produced boards were satisfied standard requirements for SWS properties (450N).

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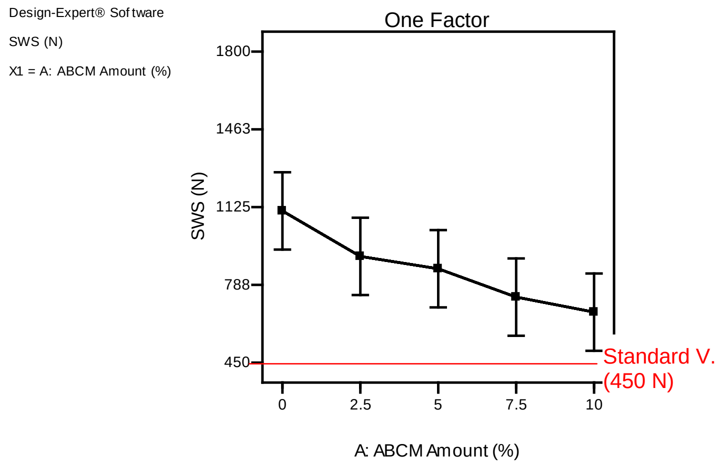

Fig. 5.
Interaction graphs of screw withdrawal strength.

The interaction graph of bending strength was shown in Fig. 6. As a result of the statistical analyses, ABCM amount had significant effect on bending strength (P=0.0002). Control group was shown higher bending strength properties than other groups containing ABCM. Based on the results, with the loading of ABCM, bending strength values decreased from control values. The highest bending strengths were observed at A0 boards produced without ABCM. Fluctuations were observed in MOR values for all boards containing ABCM. All manufactured particleboards reached standards requirements for bending strength (11MPa).

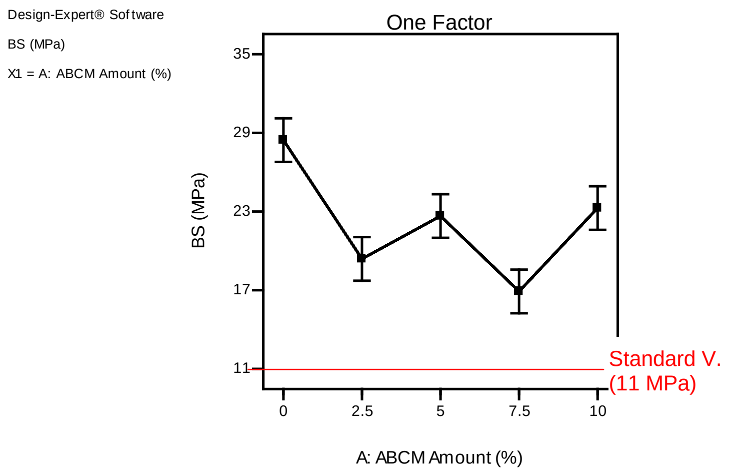

Fig. 6.
Interaction graphs of bending strength.

ABCM amount statistically had significant effect on the modulus of elasticity property (P<0.0001) as can be observed in Fig. 7. Modulus of elasticity was shown distinct decline with the first ABCM loading rate, but it was observed some variation in MOE values, with the same trend of decreasing as for MOR values. Adding ABCM, the MOE values decreased with about 27% than reference samples (without scavenger). The highest result for modulus of elasticity was obtained from A0 boards. Moreover, modulus of elasticity (MOE) values for all produced board reached standards requirements (1600MPa).

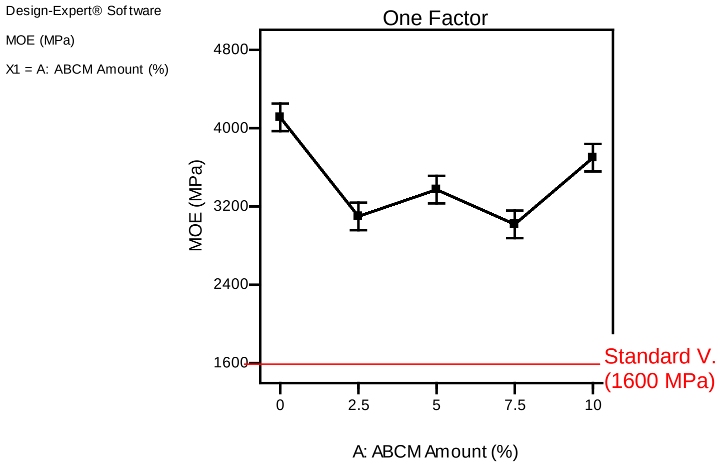

Fig. 7.
Interaction graphs of modulus of elasticity.

The surface soundness (SS) average values are presented in Fig. 8. The surface soundness values slightly increased with the rising of ABCM in the boards. ABCM amount was significantly effective on the surface soundness (P=0.0222). The best result was obtained from A10 boards. It was thought that ammonium chloride in the ABCM which used as a hardener for urea formaldehyde resin might be provided faster and better plasticization (Pizzi 1994; Pizzi and Mittal 2003; Chanda and Roy 2007) on the surface layer. It was helped to improving surface soundness properties. The images of the SS test are shown in Fig. 9. All manufactured particleboards reached standards requirements for surface soundness properties (0.80MPa).

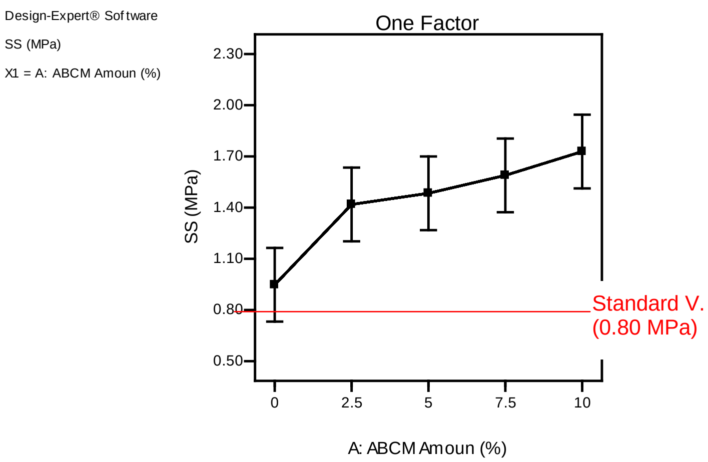

Fig. 8.
The effect of ABCM rate on the surface soundness.

Fig. 9.
Surface soundness test.

Thickness swelling and water absorption are shown in Fig. 10, Fig. 11, respectively. Although variation in values was observed on thickness swelling with the rising of ABCM amount, the results were in close range. ABCM amount had no significant effect on the thickness swelling properties (P=0.4869). All the boards manufactured with MIPW were not satisfied standards requirements (max. 15%) except A7.5 boards, but all results for all manufactured groups were close to standard requirement value. These results might be caused by lack of paraffin usage in the formulation. During commercial particleboard manufacturing paraffin (0.5-1%) is used as water repellent material to obtain better thickness swelling and water absorption properties. In this study, no water repellent was used. It was believed that usage of paraffin or other water repellent chemicals might help satisfying standard requirements or provide over the standard values for all groups. In literature, it has been reported that some rate of paraffin usage in certain proportions in manufacturing of particleboard improved thickness swelling and water absorption properties (Heebink 1967; Amthor and Böttcher 1984, Nemli et al. 2006). Water absorption properties of manufactured boards had similar trend as thickness swelling with slight increase and decrease depending on the ABCM amount. However, all manufactured groups were satisfied standard requirements (max 80%). ABCM amount had not significant effect on the water absorption properties (P=0.8903).

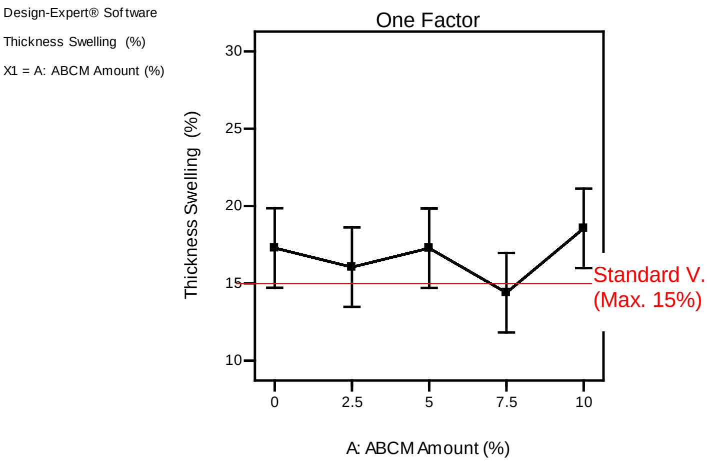

Fig. 10.
Interaction graphs of thickness swelling (at 24h).

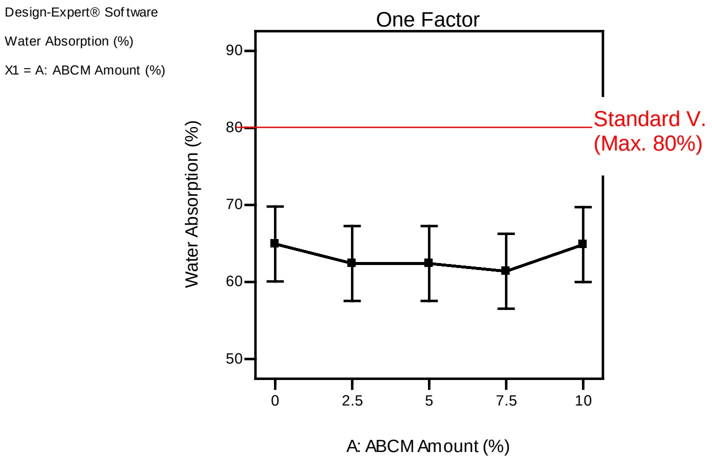

Fig. 11.
Interaction graphs of water absorption (at 24h).

CONCLUSION

The results of the study show that ABCM can be used successfully as a formaldehyde scavenger in the manufacturing of particleboards with MIPW. The experimental boards were tested and the following conclusions were reached:

1. The usage of ABCM improved the performance of boards concerning the formaldehyde content; with the increasing of ABCM rate, formaldehyde content values were reduced, Amount of ABCM mixture had significant effect on formaldehyde emission properties.
2. The impact of ABCM on bending strength, modulus of elasticity and surface soundness is found statistically meaningful ($\alpha = 0.05$), while on internal bond strength and screw withdrawal strength was not. However, the mechanical properties values met or were above standard requirements for all groups.
3. Surface soundness values were slightly increased with the rising of ABCM rate.
4. ABCM amount was not found to be statistically significant on physical properties (thickness swelling, water absorption and density).

All tested boards didn't meet the requirement for E1 class; even A10 group at max rate of ABCM had the lowest formaldehyde content. Regarding formaldehyde as an issue for the human health, it could be said that the best results were obtained from A10 group. As a result, Amine based chemical mixture (ABCM) might be used as a formaldehyde scavenger during particleboard manufacturing with the melamine impregnated paper waste, but should increase the amount of ABCM to accomplish the requirements for E1 class. Moreover, effect of more than 10% ABCM usage on the formaldehyde performance, mechanical and physical properties should be investigated. In addition, usage of paraffin or other water repellent chemicals might help to satisfy standard requirements for thickness swelling and water absorption.

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