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Dimensional Stability Properties of Medium-Density Fiberboards Produced Using Silicone-Based Chemicals

Dimenzijska stabilnost srednje gustih ploča vlaknatica proizvedenih uz upotrebu kemikalija na bazi silikona

ORIGINAL SCIENTIFIC PAPER

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ABSTRACT • The purpose of this study was to investigate the effects of two different silicon-based water-repellent chemicals (Dow Corning 87 and Xiameter PMX-200 1000cs) added to melamine urea formaldehyde glue (1.07 mol, 5% melamine additive) in different proportions (0.5%, 1.0%, 1.5% and 3.0%) on the dimensional properties (swelling-shrinkage and length extension and shrinkage) of the medium density fiberboard. In the context of the study, the dimensional stability properties of the boards were measured based on the TS EN 318:2005 standard. It was observed that, when the relative humidity of the air conditioning room increased from 65% to 85%, the swelling decreased and the extension increased, respectively, in the thickness and length directions of the boards produced with silicon-based chemicals (compared to the control board produced without using silicone-based chemicals). On the other hand, it was determined that the shrinkage in the thickness and length direction of the boards decreased when the relative humidity of the air-conditioning room was reduced from 65% to 30%. Moreover, the dimensional stability properties of medium density boards produced with silicon-based chemicals increased (resistance to moisture).

KEYWORDS: *MDF*; *hydrophobic*; *silicone-based chemical*; *water repellent*; *dimensional stability*

SAŽETAK • Cilj ove studije bio je istražiti učinke dviju različitih vodoodbojnih kemikalija na bazi silikona (Dow Corning 87 i Xiameter PMX-200 1000cs), dodanih melamin-ureaformaldehidnom ljepilu (1,07 mol, 5 % melamina) u različitim udjelima (0,5 %, 1,0 %, 1,5 % i 3,0 %) na dimenzijska svojstva srednje gustih ploča vlaknatica (bubrenje/utezanje i promjene dimenzija po duljini). Mjerenje dimenzijske stabilnosti provedeno je prema standardu TS EN 318:2005. Utvrđeno je da se s povećanjem relativne vlažnost zraka u klimatiziranoj prostoriji sa 65 na 85 % u ploča proizvedenih uz dodatak kemikalija na bazi silikona (u usporedbi s kontrolnim pločama proizvedenima bez kemikalija na bazi silikona) bubrenje u smjeru debljine smanjilo, a istezanje u smjeru duljine povećalo. Istodobno je uočeno da se utezanje ploča u smjeru debljine i duljine smanjilo kada je relativna vlažnost zraka u klimatiziranoj prostoriji smanjena sa 65 na 30 %. Štoviše, povećala se dimenzijska stabilnost srednje gustih ploča vlaknatica proizvedenih s kemikalijama na bazi silikona (otpornost na vlagu).

KLJUČNE RIJEČI: MDF; hidrofoban; kemikalija na bazi silikona; vodoodbojan; dimenzijska stabilnost

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1 INTRODUCTION

1. UVOD

Medium-density fiberboard (MDF) is a hygroscopic material like solid wood. Since it is composed of wood fibers, its dimensions (thickness and longitudinal direction) change by absorption or desorption due to changes in temperature and relative humidity of the environment (Ayrılmış and Mater, 2007; Grigsby et al., 2012; Dopico and Heroux, 2004; Xu and Winistorfer, 1995). The change in the dimensions of a board occurs as swelling/shrinkage in the thickness direction and as extension/shrinkage in the length direction. Changes in the dimensions of the boards are an important issue in terms of their end-use and the board storage areas. Therefore, various experiments have been conducted by focusing on different board production parameters, such as raw material preference, press conditions, different glue content, board density, and water-repellent chemicals.

The hygroscopic structure of wood is formed by the hydroxyl groups of polymers in the cell wall (Papadopoulos et al., 2019). It is important to ensure that these hydroxyl groups form bonds that are resistant to water and moisture and do not break easily. For this purpose, a number of studies, in which different glues and water-repellent chemicals are used and different board-forming methods are applied, have been conducted (Halligan, 1970; Mantanis and Papadopoulos, 2010a; Mantanis and Papadopoulos, 2010b). Besides the production of melamine and phenolic-containing glues, researchers have also investigated the addition of chemical substances such as water-repellent rosin and wax in order to improve the water and moisture resistance of fiber boards (Segovia et al., 2021; Moreno-Anguiano et al., 2022). As a result of these studies, water and moisture resistant boards have been produced, but completely water-resistant (unchangeable in size) boards could not be produced. However, it has been determined that long-term use of boards produced with the wax chemical causes changes (swelling- elongation) in their dimensions (Garcia et al., 2005; Halligan, 1970; Hsu et al., 1990; Press, 1990). This situation has led researchers to search for using different chemicals in board production. Various water-repellent chemicals, such as nanotechnology compounds, nanowollastonite, oils, organosilicon, and organo-silane compounds, have been tested, and boards with different levels of water-moisture resistance have been produced (De Vetter et al., 2011; Esmailpour et al., 2021; Hassani et al., 2019; Ibrahim et al., 2016; Kloeser, 2010; Mantanis and Papadopoulos, 2010a; Taghiyari et al., 2015; Wang et al., 2020).

Silicones with water-repellent properties are hydrophobic substances and have a positive effect (reduction in water and moisture absorption) on the interaction of wood-based boards with water-moisture (Donath et al., 2006; Ghosh, 2009; Aziz et al., 2021). In addition to their moisture resistance in the production of wood-based boards, they are used in a wide variety of fields such as textiles, cosmetics, wood preservation and furniture (Buyl, 2007). For the boards to remain dimensionally stable, it is necessary to add water-repellent chemicals as well as glue; the compatibility between the used water-repellent chemicals and the glue is important. In this sense, the reactive silane groups of silicon-based chemicals act as binders for both organic and inorganic components. Moreover, because of their dual reactive properties, they form bridges between inorganic and organic surfaces (cellulose- filling material) and organic polymeric matrices (e.g., rubber-thermosets), and they increase adhesion. (Materne et al., 2004; Kartal et al., 2009). These properties of silicon-based chemicals make them important in board production. In addition, their ability to bind inorganic fillers or fibers to organic glues to create or promote a stronger bond at the interface is another reason for their preference in board production. Furthermore, the chemical substances used in the production of the board do not threaten human and environmental health. In this sense, for sectors such as food industry, textile and medical applications, silicone-based chemicals that do not contain threatening elements can be safely used in production (Mai and Militz, 2004).

This study aimed to investigate the effect of Dow Corning 87 and Xiameter PMX-200 1000cs chemicals on the dimensional stability properties of medium-density fiberboard. The density of MDF boards, produced with a mixture of MUF glue and silicone-based chemicals added at different concentrations and their dimensional stability properties (swelling-shrinkage in thickness, extension-shrinkage in length direction) occurring based on the effect of the amount of chemicals, was evaluated.

2 MATERIALS AND METHODS

2. MATERIJALI I METODE

2.1 Materials

2.1. Materijali

In this study, mixed fibers produced from softwood (pine 50 %) and hardwood (beech 40 % +oak 10 %=50 %) were obtained from Çamsan Anonym Company (Sakarya, Turkey). Melamine Urea Formaldehyde (F/U: 1.07 mol 5 % melamine added) glue and ammonium chloride (glue hardener) supplied from Çamsan A.Ş. (Sakarya, Turkey) were used. Dow Corning 87 (DC) and Xiameter PMX-200 1000cs (XM) produced by Dow Corning company (Seneffe, Belgium) were used as chemicals. DC (Octyltriethoxysilane) is a liquid and white color chemical with boiling point > 35 °C and dynamic viscosity of 50 mPa·s. On the other hand, XM (Trimethylsiloxy) is a liquid and colorless chemical whose initial boiling point and boiling range are higher than 65 °C, flash point is higher than 120 °C, relative density is 0.97 and kinematic viscosity is 1000 cSt (25 °C). DC is an active (40 %) silane-based silicone glue emulsion, and its components are determined by the XRF device as given in Table 1.

 Table 1 XRF elemental analysis results of DC chemical

 Tablica 1. XRF rezultati elementarne analize DC kemikalije

Component	Solid content, %			
Komponenta	Udio suhe tvari, %			
Si	0.432			
S	0.056			
Cu	0.003			
Plastic	99.508			

The chemical name of XM is silicone and it is a linear polydimethylsiloxane. Its components are determined by XRF device as given in Table 2.

 Table 2 XRF elemental analysis results of XM chemical

 Tablica 2. XRF rezultati elementarne analize XM kemikalije

Component	Solid content, %
Komponenta	Udio suhe tvari, %
Si	25.665
S	0.052
Cu	0.008
K	0.052
Fe	0.004
Plastic	74.220

The properties of 5 % melamine added Melamine urea formaldehyde glue are presented in Table 3.

2.2 Methods

2.2. Metode

2.2.1 Board production

2.2.1. Proizvodnja ploča

The fibers supplied wet from the factory were laid out and dried in the laboratory, and the fiber lumps formed after drying were opened by pressing lightly on the sieve surface. After the opened fibers were dried in a drying oven (100 °C) until 7 % dryness, they became ready for use in board production. In order to create the fiber board, first, the glue, hardener and silicon chemi-

cal were mixed in a baker. Then the mixture was sprayed into the container in which the fibers were located, and the mixing process was carried out. After the fiber and glue mixture was homogeneously laid on the frame placed on the press board steel, manual pre-compression was performed with the help of a flat surface. After the other press-board steel was placed on the fiber mat and the thickness wedges were placed, the hot press was applied. In the context of the study, a total of 16 boards with 8 different board groups were produced. The amount of glue, fiber and hardener were calculated depending on the dry weights. The amount of glue was 20 % based on the amount of dry fiber, and the amount of hardener was used as 2 % based on dry glue. Silicon chemicals were applied at the content ratios of 0 %, 0.5 % - 1 %, 1.5 % and 3 % (w/w of fiber). The boards were formed by keeping the draft of the board in a single-layer press at 180 °C, under a pressure of 45-30 kg/ cm² for 420 seconds. Boards were produced in 300 mm \times 300 mm \times 10 mm (length \times width \times thickness) dimensions. The test procedures were started after the produced boards were conditioned at room temperature (20±2 °C, 65 % relative humidity) until reaching equilibrium moisture.

2.2.2 Board tests

2.2.2. Ispitivanje ploča

2.2.2.1 Determination of dimensional change properties of boards

2.2.2.1. Određivanje dimenzijskih promjena ploča

The test of wood based-panels determination of dimensional changes associated with changes in relative humidity (TS EN 3182005) was applied to the test samples. This test is a method based on calculation of the difference between the thickness and length of the board by keeping it in an air conditioning room (20 °C, 65 % - 85 %, 65 % - 30 %) until it reaches constant weight at different relative humidity values. Test pieces were obtained by cutting them to be (300 ± 2) mm × (50 ± 2) mm \times 10 mm (board thickness) in dimension. A total of 64 test samples were prepared by taking 8 test samples from each board group. A conditioning process consisting of three stages was applied to the board pieces separately as two sets. This process kept going until each board piece reached a constant mass. The length, thickness and mass were measured after the 2nd and 3rd conditioning process. The dimensional change in the length and thickness of the board sam-

Table 3 Properties of melamine urea formaldehyde (Çamsan Sakarya AŞ., Turkey, 2018)**Tablica 3.** Svojstva melamin-ureaformaldehida (Çamsan Sakarya AŞ., Turkey, 2018.)

Glue type	Solid matter, %	pН	Viscosity (flow time, 20 °C), s	Gel time, s	Density (20 °C), g/cm ³
Vrsta ljepila	Udio suhe tvari, %		Viskoznost (vrijeme istjecanja,	Vrijeme	<i>Gustoća (20</i> °C), g/cm ³
			<i>20</i> °C), s	<i>želiranja</i> , s	
1,16 mol MUF	60	9.11	43	48	1258

ples at different relative humidity values was calculated with the formula below.

Change in the length of the board piece:

$$\delta I_{65,85} = \frac{I_{85} - I_{65}}{I_{65}} \cdot 100 \text{ (with using results of 1 team)}$$

$$\delta I_{65,30} = \frac{I_{30} - I_{65}}{I_{65}} \cdot 100 \text{ (with using results of 2 team)}$$

Where $\delta I_{65,85}$ is relative change in length (mm/m) for the change of relative humidity from 65 % to 85 %. $\delta I_{65,30}$ is relative change in length (mm/m) for the change of relative humidity from 30 % to 65 %. I_{85} is verified length (mm) between measuring points at 20 °C temperature and 85 % relative humidity for the effect of marking used. I_{65} is verified length (mm) between measuring points at 20 °C temperature and 65 % relative humidity for the effect of marking used. I_{30} is verified length (mm) between measuring points at 20 °C temperature and 65 % relative humidity for the effect of marking used. I_{30} is verified length (mm) between measuring points at 20 °C temperature and 30 % relative humidity for the effect of marking used.

Change in the thickness of the board piece:

$$\delta t_{65,85} = \frac{t_{85} - t_{65}}{t_{65}} \cdot 100 \text{ (with using results of 1 team)}$$

$$\delta t_{65,30} = \frac{t_{30} - t_{65}}{t_{65}} \cdot 100 \text{ (with using results of 2 team)}$$

Where $\delta t_{65,85}$ is relative change in thickness for the change of relative humidity from 65 % to 85 %. $\delta t_{65,30}$ is relative change in thickness for the change of relative humidity from 30 % to 655 % t_{85} is verified thickness (mm) for the effect of marking measured at 20 °C temperature and 85 % relative humidity and used when necessary. t_{65} is verified thickness (mm) for the effect of marking measured at 20 °C temperature and 65 % relative humidity and used when necessary.

 t_{30} is verified thickness (mm) for the effect of marking measured at 20 °C temperature and 30 % relative humidity and used when necessary.

2.2.2.2 Determination of density values of boards

2.2.2.2. Određivanje gustoće ploča

The density values of the samples were determined based on the principles specified in the EN 323:1999 standard. Four test samples were used to determine the board density and the averages of the obtained results were taken. After measuring the weight, thickness and width of two edges of the test samples, which were conditioned and prepared in sample sizes of 50 mm \times 50 mm \times 10 mm, their densities were calculated with the formula below. A total of 32 test samples, four from each board, were used for the test.

$$\delta = \frac{m}{a_1 \cdot a_2 \cdot t} \cdot 10^3 (g \,/\, cm^3)$$

Where δ is density (g/cm³), *m* is air dry weight (g), a_1 and a_2 are sample width (mm), *t* is sample thickness (mm).

2.2.2.3 X-Ray Fluorescence Spectrometer 2.2.2.3. Rendgenski fluorescentni spektrometar

The contents of DC and XM chemicals were determined using an "Energy Dispersive X-ray Fluorescence Spectrometer" (Shimadzu EDX 8000, Japan). Our samples were placed in a sample cell (30 mm diameter) with film on the bottom. The amount of substance in the sample was measured by sending X-rays on it. The measurements were automatically transferred to the computer screen connected to the device. XRF (Setaş Kimya AŞ.) measurement process was done in the laboratory.

2.2.3 Statistical analysis

2.2.3. Statistička analiza

To determine the effects of the added chemicals, the dimensional stability test of the fiberboards was carried out with SPSS 22.0 software (version 29.0.0.0) by using the One Way ANOVA test at 95 % confidence interval. The mean and standard deviation values of each group were calculated, and significant differences between the groups were determined by Duncon homogeneity test.

3 RESULTS AND DISCUSSION

3. REZULTATI I RASPRAVA

3.1 Changes in board dimensions

3.1. Promjene dimenzija ploča

3.1.1 Changes in thickness

3.1.1. Promjene debljine ploča

In Table 1, swelling-shrinkage values in the thickness direction of the boards are given depending on the chemical substance concentrations used in the study.

Figures 1A and 1B show the mean thickness (swelling/shrinkage) change values of the boards produced with the addition of DC and XM chemicals when the relative humidity was increased from 65 % to 85 % and decreased from 65 % to 30 %. In both graphs, decrease in swelling/ shrinkage values of the boards in the thickness direction compared to the control board was observed depending on the chemical ratio added. As the addition amount of DC and XM chemicals increased, the changes in the thickness direction (swelling and shrinkage) of the boards decreased compared to the control board, but this change was not directly proportional to the chemical addition rate. However, changes in the form of decrease and increase in board thickness were observed.

Among the boards produced by adding XM chemical, the lowest increase in thickness was obtained in the board in which XM was used at a rate of 1 % (4.45 %). The lowest increase in thickness in the boards produced by adding DC chemical was determined in

Dimensional stability / Dimenzijska stabilnost							
% Cons. DC	Density, g/cm³ <i>Gustoća</i> , g/cm ³	$\delta t_{(65,85)}$	$\delta t_{(65.30)}$	% Cons. XM	Density, g/cm³ <i>Gustoća</i> , g/cm ³	$\delta t_{(65.85)}$	$\delta t_{(65,30)}$
0	762	5.14(0.16)* °	-2.45 (0.08) a	0	762	5.14 (0.16)*d	-2.45 (0.08) ^a
0.5	760	5.16 (0.13)°	-1.85 (0.13) °	0.5	764	4.58 (0.12) b	-2.13 (0.14) ^b
-	-	-	-	1	757	4.45 (0.39) a	-1.94 (0.01) °
1.5	801	4.34 (0.39) a	-1.67 (0.27) ^d	1.5	758	4.86 (0.41) °	-2.06 (0.06) ^b
3	753	4.89 (0.31) ^b	-2.32 (0.9) ^b	3	761	4.70 (0.06) ^b	-1.93 (0.19) °

Table 4 Swelling-shrinkage properties in terms of thickness direction of MDF boards **Tablica 4.** Debljinsko bubrenje i utezanje MDF ploča

*Numbers in parentheses are standard deviations. The sample with 0 % concentration is the control board. Letters represent each homogenous subset analyzed with Duncan test.

*Brojevi u zagradama standardne su devijacije. Uzorak s 0 % koncentracije kontrolna je ploča. Slova predočuju svaki homogeni podskup analiziran Duncanovim testom.



Figure 1 (A) Change in thickness of the boards produced with XM chemical, (swelling/shrinkage), (B) change in thickness of the boards produced with DC chemical (swelling/shrinkage)

Slika 1. (A) Promjena debljine ploča proizvedenih s XM kemikalijom (bubrenje/utezanje), (B) promjena debljine ploča proizvedenih s DC kemikalijom (bubrenje/utezanje)

the board produced by adding 1.5 % DC. When both chemicals were compared, it was observed that the lowest thickness increase (4.34 %) occurred in the board produced by using DC chemical at a rate of 1.5 %. The fact that the swelling and shrinkage values in

the thickness direction do not decrease regularly with the increase in the amount of chemical addition is attributed to the chemical-fiber bond. In other words, the fact that the swelling and shrinkage values did not change regularly, although the amount of fiber used in the production of the boards was the same and the chemical amount added increased, was attributed to the fact that the number of bonds did not increase between chemical and fiber.

When the board density values were examined, the lowest change of thickness (swelling-shrinkage) was observed in the board with the highest density value (board produced by adding 1.5 % DC). It is known that as the board density value increases, the change in the dimensions of the boards increases when the boards receive water and moisture (Akbulut and Ayrılmış, 2008). However, in our study, the lowest swelling and shrinkage amount was determined in the thickness direction of the board produced with 1.5 % DC chemical, which is high board density (at relative humidity change). As the board density increased, more fiber-chemical bonds were formed, and because of this, less swelling occurred in the thickness direction of the board. When the test boards were compared with the control board, it was determined that there was a 13.42 % (calculated based on the swelling value of control board) decrease in the thickness value of the board to which 1 % XM was added and a 15.56 % (calculated based on the swelling value of control board) decrease in the thickness value of the board to which 1.5 % DC was added. When this comparison was made in terms of shrinkage, a 21.22 % (calculated based on the shrinkage value of control board) decrease was obtained in the board to which 3 % XM was added, and a decrease of 31.83 % (calculated based on to the shrinkage value of control board) in the board to which 1.5 % DC was added. In the boards to which DC and XM chemicals were added, less contraction (shrinkage) was obtained in the thickness direction.

Reducing the water absorption and swelling rates of the boards depends on the formation of strong bonds by the free -OH groups in the fiber and the fact that these bonds are not broken. It has been stated in studies that such strong fiber bonds can be achieved with cross-linking chemicals (Mamiński et al., 2020; De Vetter, 2009). The reactive silane groups in the content of silicon-based chemicals bond with the hydroxyl groups of the fiber, and although the hydrogen bonds formed are sensitive to hydrolysis, they turn into covalent bonds by heat (Xie et al., 2010). These covalent bonds are stable bonds. Although there are functional organic and alkoxy groups in the silane-containing chemical, these groups provide adhesion between the fiber and the polymer matrix (Goyal, 2006). Organic groups interact with the polymer, while silanol groups improve the interfacial properties by creating covalent bonds on the inorganic surface (Nurazzi et al., 2021). In addition, alkoxy groups react with the hydroxyl groups of the fiber under humidity conditions and form silanols (Agrawal et al., 2000). While the hydroxyl groups of the fiber and the reactive silane groups bond together, the free silanols form a stable Si-O-Si bond by bonding with each other. These bonds form solid polysiloxane structures because of condensation reaction occurring with the effect of heat on the fiber surface. These polysiloxane layers prevent the water absorption of the fibers close to the surface, which reduces the likelihood of bonding with the hydroxyl groups of the fiber. The presence of these polysiloxane layers explains the reduction of thickness (swellingshrinkage) of the boards produced.

In the study, it was determined that the increase in thickness of the produced boards was greater than the decrease in thickness (well-known hysteresis phenomenon). The bond established between chemical-fiber, chemical-glue and fiber-glue is broken if the moisture value of the board is increased or decreased. If the broken bonds are re-established (bond formed between fiber and moisture) bonds with the moisture taken by the board, it will cause differences in board thickness. The increase in the thickness of boards has been attributed to the relaxation of the compressive stresses in the board, the hygroscopic swelling of the fibers, and the deterioration of the bond between fibers. In this context, it is expected that the decrease in the thickness of the boards will be less than the increase in the thickness. (Ayrılmış and Mater, 2007). Rather than the hygroscopic nature of the boards, this event is due to the deterioration of the compression caused by the press pressure during the board production. When the boards absorb or desorb moisture, their thickness changes with the broken bonds. If the boards desorb moisture again after absorbing it, they cannot reach their original dimensions. If the board gets moisture, new bonds formed between moisture with fiber, glue and chemical affect the board thickness. The fact that the free -OH groups of the fiber form bonds with water causes the board to swell. These bonds will affect the thickness of the board, but the board will not reach its original state. It is thought that the same has occurred in the boards produced in the study.

3.1.2 Changes in length

3.1.2. Promjene duljine ploča

In Table 5, extension-shrinkage values in the length direction of the boards are shown based on the chemical substance concentrations used in the study.

Figures 2A and 2B show the average length (extension/shrinkage) change values that occurred when the relative humidity value of the boards produced with the addition of DC and XM chemicals at different rates changes (65 % - 85 % or 65 % - 30 %). It was observed that, as the relative humidity increased, an elongation occurred in the length direction of boards produced with DC and XM compared to the control board. It was deter-

Dimensional stability / Dimenzijska stabilnost							
% Cons. DC	Density, g/cm³ <i>Gustoća</i> , g/cm ³	$\delta t_{\scriptscriptstyle (65,85)}$	$\delta t_{(65.30)}$	% Cons. XM	Density, g/cm³ <i>Gustoća</i> , g/cm ³	$\delta t_{(65.85)}$	$\delta t_{(65,30)}$
0	762	1.79 (0.12)*a	-2.92 (0.04) ^a	0	762	1.79 (0.12) ^a	-2.92 (0.04) ^a
0.5	760	2.43 (0.11) ^b	-2.69 (0.05) ^b	0.5	764	2.41 (0.02)°	-2.72 (0.09) ^b
-	-	-	-	1	757	2.52 (0.01) ^d	-2.59 (0.04) ^d
1.5	801	2.39 (0.19) ^b	-2.56 (0.02)°	1.5	758	2.19 (0.09) ^b	-2.69 (0.09) ^b
3	753	2.42 (0.19) ^b	-2.55 (0.01) ^c	3	761	2.26 (0.04) ^b	-1.93 (0.07)°

Table 5 Extension-shrinkage properties in length direction of MDF boards **Tablica 5.** Produljenje i skupljanje MDF ploča po duljini

*Numbers in parentheses are standard deviations. The sample with 0 % concentration is the control board. Letters represent each homogenous subset analyzed with Duncan test.

*Brojevi u zagradama standardne su devijacije. Uzorak s 0 % koncentracije kontrolna je ploča. Slova predočuju svaki homogeni podskup analiziran Duncanovim testom.





Slika 2. (A) Promjena duljine ploča proizvedenih s XM kemikalijom (produljenje/utezanje), (B) promjena duljine ploča proizvedenih s DC kemikalijom (produljenje/utezanje)

mined that the shrinkages in the length direction of the boards when the relative humidity decreased were less compared to the control group board. The lowest length shrinkage was observed in the board sample to which the XM chemical was added at a rate of 3 %.

The graph shows that the change in the density values of the boards produced using XM chemical is

lower than the change in density values of the boards produced using DC. In the case of the decrease in the relative humidity, the lowest shrinkage value (-1.93 mm/m) was determined on the board produced with 3 % XM addition. On the other hand, in the case of the decrease in the relative humidity in the boards produced with DC addition, the shrinkage in the longitudinal direction decreased, and the lowest value was determined in the board produced with the 3 % DC addition (-2.55 mm/m).

The reason why the change in the direction of length is high in the boards produced with DC and XM chemicals is that, when the board absorbs moisture from the surface, the other Si-OR bonds, formed next to the Si-OH bond groups created by free OH groups and silanols in the fiber, easily hydrolyzed by moisture (Buyl, 2007). Based on this, it can be said that the bonds deteriorating in the presence of moisture cause the stress in the board to relax and increase the distance between the fibers in the length direction, and this is also effective in the length extension of the board sample.

According to Özen (1975), the work of the boards (desorption- absorption of water) in the longitudinal direction is the opposite of the work in the thickness direction. If the board loses moisture, the length change in the longitudinal direction is high. The boards absorb moisture at high relative humidity; however, they do not release all the moisture they take in when the humidity drops, while more shrinkage is seen on them. This has been attributed to the hysteresis of the boards (Ganev, 2002; Ganev et al., 2005). Although the extension in the length direction does not directly reflect the free expansion of the individual particles, it is a value that occurs depending on the fiber orientation and the degree of limitation of the bound expansion based on the direction of expansion of individual particles (Xu and Suchsland, 1996). In the board production performed under laboratory conditions, it is not possible to ensure homogeneous and regular distribution of fibers. In this case, the change in the dimensions of the board with the change in humidity and density is expected. This situation was also observed in the boards produced in this study.

Although silicones are chemicals that provide hydrophobic properties, diffusion of gases into silicones occurs due to their high free volume and solubility. Silicone surfaces cannot be wetted with water, but they cannot prevent the transition of oxygen and nitrogen (Colas, 2005). Based on the results obtained, it can be said that the cross-links formed because of the bonding of silane groups in silicone-based chemicals with the free OH groups of the fiber are effective on the less moisture absorption of the boards and less expansion and contraction in their dimensions.

It was determined that the changes in the thickness and length direction of the boards were not at the same rate when the boards produced with XM and DC chemicals were exposed to same climate conditions. This difference can be explained by the fact that both chemicals have different properties. DC chemical is a white colored liquid emulsified resin containing 40 % active silane, while XM chemical is an oily, transparent-colored and intense chemical containing polydimethylsiloxane groups. It was thought that the fact that the DC chemical was liquid causes it to reach the fiber and to obtain more bonding opportunities. XM chemical, on the other hand, was thought to form polysiloxane layers on the fiber surface due to its dense and oily chemical structure (Xie et al., 2010). There was no regular (systematic) change in the change values in the thickness (swelling-shrinkage) and length (extension-shrinkage) directions when the boards absorbed or lost moisture due to the increase in the DC and XM chemical substance ratios. It was observed that there were increases and decreases in the change values (swelling-shortening or extension-shortening) of the boards depending on the additional chemical ratios. This situation suggested that the silanes in the chemicals could not bond with all the -OH groups of the fiber and that at the same time, a bond was formed between fiber-fiber and fiber-glue. In addition, it was concluded that the increase in the amount of chemical substance added without changing the fiber amount did not increase the number of silane-fiber bonds. It is thought that the amount of silane that cannot bond with the increase in the amount of chemical substance may increase, and these silanes may remain clustered on the surface of the fiber or on the cell wall (Xie et al., 2010). In addition, it is seen that the -Si contents of both chemicals are different when the contents of DC and XM chemicals are examined (Table 1 and Table 2). In the study, it was observed that the -Si ratio in the DC chemical was lower than the -Si ratio in the XM chemical. The high amount of -Si in the chemical added at the same rate was interpreted as the fact that the number of bonds formed with the -OH groups of the fiber could be different. In this case, it was expected that the changes in the dimensions of the boards against humidity would be different and the results were obtained as expected. In addition, it was shown that the high plastic content of both chemicals was effective on boards to gain moisture resistance. The results revealed that the minimum change values were in the dimensions of boards produced with 1.5 % DC and 1 % - 1.5 % XM.

4 CONCLUSIONS

4. ZAKLJUČAK

In this study, the dimensional changes in the thickness (swelling/shrinkage) and length (extension/ shrinkage) directions of the medium-density fiber boards produced with the mixture of Muf glue and two different silicon-based chemicals in different ratios were investigated and compared to the control board.

The results of the study are summarized below:

When the relative humidity of the environment, where the produced board samples were located, were increased from 65 % to 85 %, a slight decrease in swelling values in the thickness direction was observed compared to the control board (silicone was not added). On the other hand, it was determined that the shrinkage values of the boards decreased in the thickness direction when the relative humidity of the environment, where the boards were located, was reduced from 65 % to 30 %.

It was observed that, when the relative humidity of the environment increased for the board samples produced with DC and XM, the length extension occurred in them (65 % - 85 %). The extension values in the length direction of the board samples produced with the addition of DC at different rates were similar to each other. The lowest extension value in the length direction was determined in the board produced with 1.5 % XM. The study revealed that the shrinkage values in the length direction decreased when the relative humidity of the environment decreased (65 % - 30 %) in all board samples produced. When the effect of XM and DC chemicals on the board length extension-shrinkage was compared, it was found that the length shrinkage of the boards produced with XM (3 %) was less pronounced.

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5 REFERENCES

5. LITERATURA

- Agrawal, R.; Saxena, N. S.; Sharma, K. B.; Thomas, S.; Sreekala, M. S., 2000: Activation energy and crystallization kinetics of untreated and treated oil palm fibre reinforced phenol formaldehyde composites. Materials Science and Engineering: A, 277: 77-82. https://doi. org/10.1016/S0921-5093(99)00556-0
- Akbulut, T.; Ayrılmış, N., 2008: Fundamental factors affecting the development of warp in MDF and particleboard. Journal of the Faculty of Forestry Istanbul University, 58 (2): 81-98 (in Turkish).
- Ayrılmış, N.; Mater, J., 2007: Effect of panel density on dimensional stability of and high density fiberboard. Journal of Materials Science, 42: 8551-8557. https://doi. org/10.1007/s10853-007-1782-8
- Aziz, T.; Ullah, A.; Fan, H.; Jamil, M. I.; Khan, F. U.; Ullah, R.; Iqbal, M.; Ali, A.; Ullah, B., 2021: Recent progress in silane coupling agent with its emerging applications. Journal of Polymers and the Environment, 29: 3427-3443. https://doi.org/10.1007/s10924-021-02142-1
- Buyl, de F., 2007: Silicones in Industrial Applications, Organo-Functional Silanes, Dow Corning Europe SA, Seneffe (Belgium).
- 6. Colas, A., 2005: Silicones: Preparation, Properties and Performance, Dow Corning, Life Sciences.
- De Vetter, L., 2009: Organosilicon compounds as potential wood protecting agents. PhD Thesis, Ghent University, Faculty of Bioscience Engineering, Ghent, Belgium, pp:153-172.

- De Vetter, L.; Van den Bulcke, J.; Van Acker, J., 2011: Envelope treatment of wood based materials with concentrated organosilicons. Europe Journal of Wood Product, 69: 397-406. https://doi.org/10.1007/s00107-010-0448-4
- Donath, S.; Militz, H.; Mai, C., 2006: Creating water repellent effects on wood by treatment with silanes. Holzforschung, 60: 40-46. https://doi.org/10.1515/ HF.2006.008
- Dopico, G. P.; Heroux, L. G., 2004: Dimensional stability of particleboard and MDF for use as substrates for lamination. In: TAPPI Decorative and Industrial Laminates Symposium.
- EN 323:1999 Wood Based panels Determination of density.
- Esmailpour, A.; Taghiyari, H. R.; Majidi, R.; Babaali, S.; Morrell, J. J.; Mohammadpanah, B., 2021: Effects of adsorption energy on air and liquid permeability of nanowollastonite-treated medium-density fiberboard. IEEE Transactions on Instrumentation and Measurement, 70, 1-8. https://doi.org/10.1109/TIM.2020.3009355
- Ganev, S., 2002: Modeling of the hygromechanical warping of medium density fiberboard. PhD Thesis, Forestry Faculty, University of Laval, Canada, p 180.
- Ganev, S.; Cloutier, A.; Beauregard, R.; Gendron, G., 2005: Linear Expansion and Thickness Swell of MDF as a Function of Panel Density and Sorption State. Wood and Fiber Science, 37 (2): 327-336.
- Garcia, R. A.; Cloutier, A.; Riedl, B., 2005: Dimensional stability of MDF panels produced from fibres treated with maleated polypropylene wax. Wood Science and Technology, 39: 630-650. https://doi.org/10.1007/ s00226-005-0028-7
- Ghosh, S. C., 2009: Wood modification with functionalized polydimethylsiloxanes. PhD Thesis, University of British Columbia, Vancouver.
- Goyal, S., 2006: Silanes: chemistry and applications. The Journal of Indian Prosthodontic Society, 6 (1): 14-18. https://doi.org/10.4103/0972-4052.25876
- Grigsby, W.; Thumm, A.; Carpenter, J., 2012: Fundamentals of MDF Panel dimensional stability: Analysis of MDF high- density layers. Journal of Wood Chemistry and Technology, 32 (2): 149-164. https://doi.org/10.1080 /02773813.2011.624667
- Halligan, A. F., 1970: A review of the swelling in particleboard. Wood Science Technology, 4: 301-312. https://doi.org/10.1007/BF00386406
- Hassani, V.; Taghiyari, H. R.; Schmidt, O.; Maleki, S.; Papadopoulos, A. N., 2019: Mechanical and physical properties of oriented strand lumber (OSL): The effect of fortification level of nanowollastonite on UF glue. Polymers, 11 (11): 1884. https://doi.org/10.3390/polym11111884
- Hsu, W. E.; Melanson, R. J.; Kozak, P. J., 1990: The effect of wax type and content on waferboard properties. In: Proceedings of 24th international particleboard composites symposium. Washington State University, Pullman, WA, pp. 85-96.
- Ibrahim, Z.; Ahmad, M.; Aziz, A. A.; Ramli, R.; Jamaludin, M. A.; Muhammed, S.; Alias, A. H., 2016: Dimensional stability properties of medium density fibreboard (MDF) from treated oil Palm (*Elaeis guineensis*) empty fruit bunches (EFB) fibres. Open Journal of Composite Materials, 6: 91-99. https://doi.org/10.4236/ ojcm.2016.64009

- Kartal, S. N.; Yoshimura, T.; Imamura, Y., 2009: Modification of wood with Si compounds to limit boron leaching from treated wood and to increase termite and decay resistance. International Biodeterioration & Biodegradation, 63: 187-190. https://doi.org/10.1016/j.ibiod.2008.08.006
- 24. Kloeser, L., 2010: Organofunctional silanes as formaldehyde free adhesives for fiberboards. In: Proceedings of the International Convention of Society of Wood Science and Technology and United Nations Economic Commission for Europe – Timber Committee, October 11-14, Geneva, Switzerland.
- Mai, C.; Militz, H., 2004: Modification of wood with silicon compounds. Treatment systems based on organic silicon compounds – A Review. Wood Science and Technology, 37: 453-461. https://doi.org/10.1007/s00226-004-0225-9
- Mamiński, M. L.; Trzepalka, A.; Auriga R.; H'Ng, P. S.; Chin, K. L., 2020: Physical and mechanical properties of thin high density fiberboard bonded with 1,3-Dimethylol-4,5-Dihydroxyethyleneurea (DMDHEU). The Journal of Adhesion, 96 (7): 679-690. https://doi.org/10.1080/00 218464.2018.1500280
- 27. Mantanis, G. I.; Papadopoulos, A. N., 2010a: Reducing the thickness swelling of wood based panels by applying a nanotechnology compound. European Journal of Wood and Wood Products, 68: 237-239. https://doi.org/10.1007/ s00107-009-0401-6
- Mantanis, G. I.; Papadopoulos, A. N., 2010b: The sorption of water vapour of wood treated with a nanotechnology compound. Wood Science and Technology, 44 (3): 515-522. https://doi.org/10.1007/s00226-010-0326-6
- 29. Materne, T.; Buyl, F.; Witucki, G. L., 2004: Organosilane technology in coating applications: Rewiev and perspective. Dow Corning Corporation.
- Moreno-Anguiano, O.; Cloutier, A.; Rutiaga-Quiñones, J. G.; Wehenkel, C.; Rosales-Serna, R.; Rebolledo, P.; Carrillo-Parra, A., 2022: Use of Agave durangensis bagasse fibers in the production of wood-based medium density fiberboard (MDF). Forests, 13 (2): 271. https:// doi.org/ 10.3390/f13020271
- 31. Nurazzi, N. M.; Shazleen, S. S.; Aisyah, H. A.; Asyraf, M. R. M.; Sabaruddin, F. A.; Mohidem, N. A.; Norrrahim, M. N. F.; Kamarudin, S. H.; Ilyas, R. A.; Ishak, M. R.; Abdullah, N. Nor N. M., 2021: Effect of silane treat-

ments on mechanical performance of kenaf fibre reinforced polymer composites: a review. Functional Composites and Structures, 3: 045003. https://doi.org/10.1088/2631-6331/ac351b

- Özen, R., 1975: Lif Levhanın Fiziksel Ve Mekanik Özellikleri Ve Bunlara Tesir Eden Faktörler. İstanbul Üniversitesi Orman Fakültesi Dergisi, XXV, II.
- Papadopoulos, N. A.; Bikiaris, D. N.; Mitropoulos, A. C.; Kyzas, G. Z., 2019: Nanomaterials and chemical modifications for enhanced key wood properties: A review. Nanomaterials, 9: 607. https://doi.org/10.3390/nano9040607
- Press, W. A., 1990: Wax: types and applications. In: Proceedings of the NPA resin and blending seminar. National Particleboard Association, Gaithersburg, Maryland, USA, pp 29-34.
- 35. Segovia, F.; Blanchet, P.; Essoua, G. G. E.; 2021: Potential of the crude glycerol and citric acid mixture as a binder in medium-density fiberboard manufacturing. European Journal of Wood and Wood Products, 79 (5): 1141-1151. https://doi.org/10.1007/s00107-021-01719-w
- Taghiyari, H. R.; Karimi, A.; Tahir, P. M., 2015: Organosilane compounds in medium density fiberboard: physical and mechanical properties. Journal of Forestry Research, 26 (2): 495-500. https://doi.org/10.1007/ s11676-015-0033-0
- ***TS EN 318:2005 Wood based panels Determination of dimensional changes associated with changes in relative humidity.
- Wang, Q.; Zhang, Y.; Liang, W.; Wang, J.; Chen, Y., 2020: Effect of silane treatment on mechanical properties and thermal behavior of bamboo fibers reinforced polypropylene composites. Journal of Engineered Fibers and Fabrics, 15: 1-10. https://doi. org/10.1177/1558925020958195
- Xie, Y.; Hill, C. A. S.; Xiao, Z.; Militz, H.; Mai, C., 2010: Silane coupling agents used for natural fiber/polymer composites: A review. Composites, Part A: Applied Science and Manufacturing, 41: 806-819. https://doi. org/10.1016/j.compositesa.2010.03.005
- Xu, W.; Winistorfer, P. M., 1995: A procedure to determine thickness swell distribution in wood composite panels. Wood and Fiber Science, 27 (2): 119-125.
- Xu, W.; Suchsland, O., 1996: Linear expansion of wood composites: A mode. Wood and Fiber Science, 29 (3): 272-281.

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