Evaluation of Styrene-based Copolymers and Glutaraldehyde for Medium-density Fiberboard Production

Derya Ustaömer,[a,*] Esat Gümüşkaya,[a] and Mehmet Erel,[b]

The performance of medium density fiberboard (MDF) with styrene-based copolymers and glutaraldehyde was evaluated. Styrene/n-butyl acrylate (SBA), styrene maleic anhydride (SMA), and glutaraldehyde (GA) were tested at 1%, 2.5%, and 5% levels. Surface roughness parameters, mechanical and physical properties, and formaldehyde emission values were evaluated. Two different methods were used for panel preparation: surface application and mixing with UF resin. Different trends were observed depending on chemical types, chemical concentrations, and application methods. The surface roughness parameters (R_a, R_q, R_z) decreased with the application methods and chemicals used. The smoothest surfaces were obtained from groups with the chemicals compared to control groups. The surface application method yielded the most favorable results. The thickness swelling (TS) and water absorption (WA) values generally showed slight improvements, and better results were obtained with the UF-mixing method. Mechanical properties such as internal bond strength (IB), modulus of rupture (MOR), and modulus of elasticity (MOE) showed variations depending on the experimental parameters. In general, higher values were obtained for both application methods compared to control values. Free formaldehyde emission values were notably reduced with the UF-mixing method. In general, the use of SBA, SMA, and GA chemicals contributed to lower formaldehyde emission values.

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Keywords: Styrene maleic anhydride; Styrene/n-butyl acrylate; Glutaraldehyde; MDF

Contact information: a: Department of Forest Industry Engineering, Karadeniz Technical University, 61080, Trabzon, Türkiye; b: Çamsan Ordu Wood Company RD Center, Ordu-Türkiye;
** Corresponding author: uderya@ktu.edu.tr*

INTRODUCTION

Wood based composites are employed in numerous application areas for structural and non-structural purposes across different product groups that include furniture components, various panels for both interior and exterior uses, and structural support elements in buildings (Stark *et al.* 2010). Especially, both composite panel types (fiberboard and particleboard) are widely used in the cabinet and furniture industries to manufacture various products such as laminate flooring, moldings, and veneered panels (Hiziroglu and Suzuki 2007). Therefore, it is important to improve the properties of these panels intended for use in various applications.

In most wood products, various additives are employed to provide these products with different properties and improve their characteristics. These include preservatives used to protect the wood against fire or biological degradation. In addition, non-wood materials can be used to overcome the weak properties of the wood material and enhance product performance (Sandberg 2016).

Among various known protein cross-linking agents, glutaraldehyde (GA) has found the widest application in many use fields. It is one of the most effective and popular reagents used to chemically cross-link proteins (Migneault *et al*. 2004). GA is a dialdehyde. In principle, this compound is capable of reacting with four hydroxyl groups of cell wall polymers. Therefore, it can be employed as a cross-linking agent to change wood properties (Xiao *et al.* 2010a). Several studies have been conducted to determine the effects of glutaraldehyde (GA) used for various purposes either alone or alongside other chemicals for wood (Xiao *et al.* 2010a,b; Xie *et al.* 2011; Xiao *et al*. 2012a,b) and wood based panels such as particleboard (Mamiński *et al*. 2008; Akinyemi *et al.* 2019), plywood (Wu *et al.* 2017; Xi *et al*. 2021) and fiberboard (Ji *et al*. 2017, 2018; Liu *et al*. 2021).

Styrene-based copolymers are high-performance thermoplastic elastomers. These copolymers have a wide range of applications and are used in both manufacturing and industrial applications (Afzal *et al.* 2017). Styrene maleic anhydride (SMA), a chemically reactive copolymer with certain functional groups, is formed by the copolymerization of styrene with maleic anhydride (Sarı *et al.* 2008). One of the uses of SMA is its application for plastic composites. These SMA-plastic composites are especially employed in the automotive industry and various applications in the field of engineering (Zor *et al.* 2016). Several research studies have been conducted in order to evaluate and to investigate the effect of styrene-based copolymers such as SMA for composites (Simonsen *et al*. 1998; Poletto 2016; Sommerhuber *et al.* 2016; Zor *et al*. 2016, 2018).

This study investigated the impact of styrene-based copolymers (styrene/n-butyl acrylate and styrene maleic anhydride) and glutaraldehyde applied using two different methods on the surface properties, physical properties, mechanical properties and formaldehyde emission content of MDF panels. The aim of the study was to improve the MDF panel surfaces for surface treatments, minimize the amount of sanding waste, and ultimately reduce production costs. The study also aimed to determine the effects on the mechanical and physical properties of the MDF panels, while reducing the free formaldehyde content in the panels.

EXPERIMENTAL

Materials

In this study, commercial fibers (70% hardwood-30% softwood), urea formaldehyde supplied from Çamsan Ordu Wood Company (Türkiye), a hardener and chemicals (styrene/n-butyl acrylate, styrene maleic anhydride and glutaraldehyde) were used for panel production.

Experimental Design

The experimental design (panel groups and application methods) used in this study is represented in Table 1.

Table 1. Panel Groups and Application Methods

Panel Groups/ Surface Application	Panel Groups/ Mixture with UF
1 % SBA-surface*	1 % SBA-UF/mixt.*
2.5% SBA-surface	2.5% SBA-UF/mixt.
5 % SBA-surface	5 % SBA-UF/mixt.
1 % SMA-surface	1 % SMA-UF/mixt.
2.5% SMA-surface	2.5% SMA-UF/mixt.
5% SMA-surface	5% SMA-UF/mixt.
1% GA-surface	1% GA-UF/mixt.
2.5% GA-surface	2.5% GA-UF/mixt.
5% GA-surface	5% GA-UF/mixt.
(Control 1) *surface-water application (Control 2) without surface-water application	

SBA: Styrene/n-*butyl* acrylate; SMA: Styrene maleic anhydrite; GA: Glutaraldehyde
Control 1*: This group was only used to evaluate for surface properties

In this study, the solutions of styrene-based copolymers and glutaraldehyde were prepared at different chemical concentrations (1%, 2.5%, and 5%) and these chemicals were applied for fiberboard production process using two different methods:

(1) Surface application before hot pressing: Styrene n-butyl acrylate (SBA), styrene maleic anhydride (SMA) and glutaraldehyde (GA) were selected at concentrations of 1%, 2.5%, and 5% based on the solid content of the resin and dissolved in 50 mL of water. Afterwards, 25 mL of this solution was sprayed under the panel mat and while the remaining 25 mL was sprayed on top of the panel mat.

(2) Mixing into UF resin: The chemicals (SBA, SMA, and GA) were mixed into the UF resin at concentrations of 1%, 2.5%, and 5% based on the solid content of resin. Then, the mixtures of UF resin (13%) at the solid-content ratio of 58%, chemicals and a hardener were sprayed onto the fibers and the panel mats were formed.

Panel Preparation

After application methods, the pressing stage of panel mats was carried out using a hot press (Cemilusta Hot Press; Cemilusta Wood Working Machinery Ind. Inc., İstanbul, Türkiye) at a temperature of 180 °C for 8 min. For each group, three panels measuring 30×30×1 cm were produced with a density of 0.65 to 0.72 g/cm³. These panels were kept in a climate-controlled room for conditioning process and then, were dimensioned for selected tests. Additionally, two control panel groups (with water-surface application, without water-surface application) were produced, and Control 1 (with water-surface application) were used to evaluate the only surface properties.

Surface Roughness Measurements

The surface roughness measurements of MDF samples were performed using Mitutoyo Surftest SJ-210 surface roughness instrument according to EN ISO 21920-2(2022) standard. The parameters R_a, R_q, *and* R_z were measured to determine surface roughness of MDF panel samples.

Physical Properties

The physical properties such as thickness swelling (TS) and water absorption (WA) for 2 to 24 h of MDF samples were carried out according to the EN 317 (1993) standard.

Mechanical Properties

Internal bond strength (IB), modulus of rupture (MOR), modulus of elasticity (MOE) of the MDF samples were carried out according to related standards EN 310 (1993) and EN 319 (1993).

Formaldehyde Emission

The formaldehyde emission amounts of MDF samples were assessed using extraction method, also known as perforator method, according to EN ISO 12460-5 (2015) standard.

RESULTS AND DISCUSSION

Surface Roughness Measurements

The surface roughness parameters R_a, R_q, and R_z of MDF samples, depending on the application methods, are presented in Figs. 1, 2, and 3.

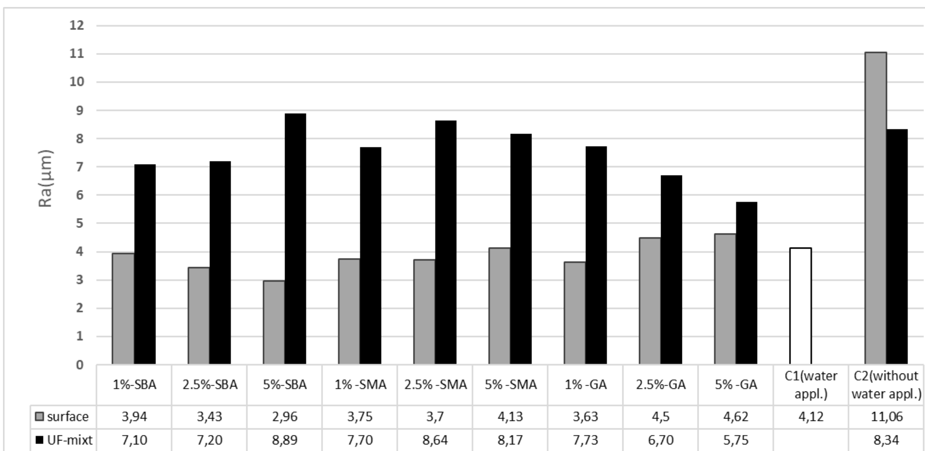


Fig. 1. R_a values of MDF samples

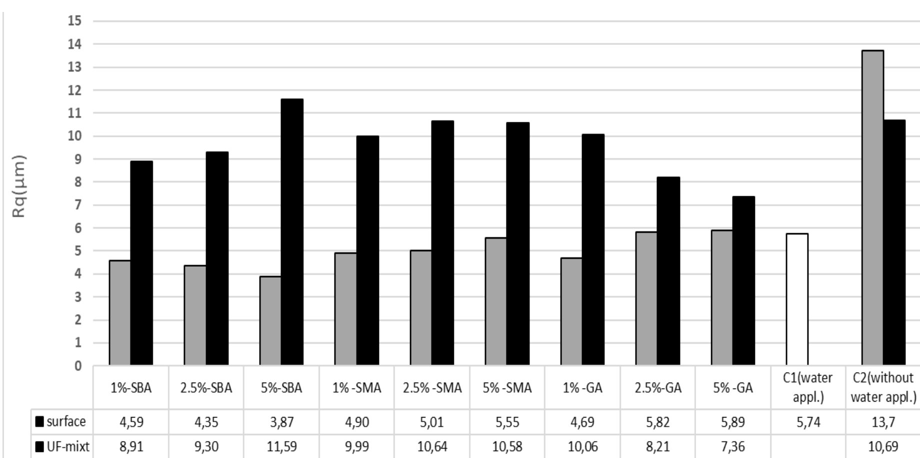


Fig. 2. R_q values of MDF samples

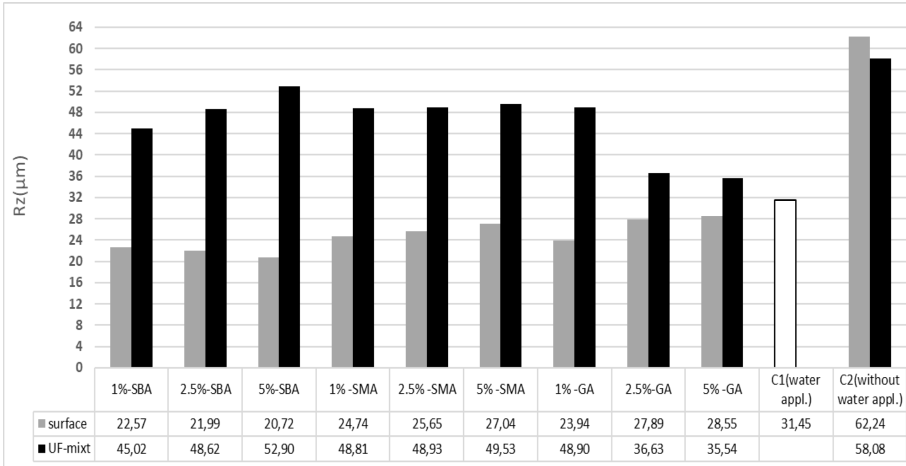


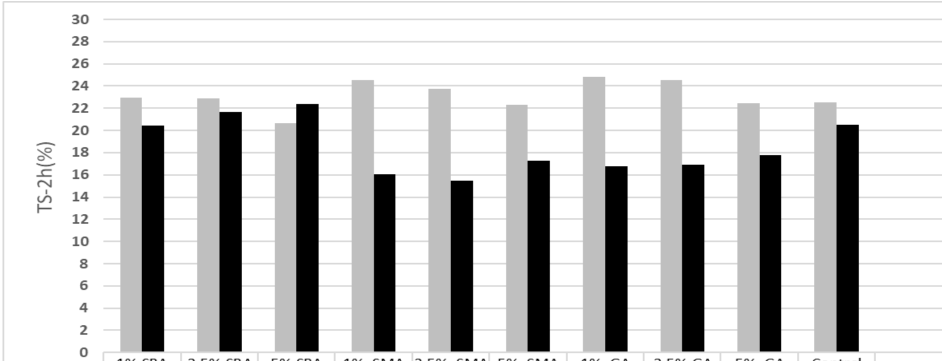
Fig. 3. R_z values of MDF samples

As can be seen from Figs. 1, 2, and 3, the surface roughness parameters (R_a, R_q, and R_z) of MDF samples varied depending on the application methods, the chemical types, and the chemical concentrations. In some groups, the surface roughness values decreased slightly with the increasing chemical concentrations, while in some groups the values increased slightly. These values ranged from 2.96 µm to 11.1 µm (R_a), 3.87 µm to 13.7 µm (R_q), and 20.7 µm to 62.2 µm (R_z) for the groups with surface application, whereas these values ranged from 5.75 µm to 8.89 µm (R_a), 7.36 µm to 11.6 µm (R_q), and 35.5 µm to 58.1 µm (R_z) for the groups with UF-mixture. All MDF panel groups for both application methods exhibited lower surface roughness values than their control groups. Especially, the best R_a, R_q, and R_z values were obtained from SBA groups with surface application as compared to both control groups (with and without surface-water application). The smoothest surfaces were obtained from 5% SBA group with surface application among the other groups. The highest surface roughness values were obtained from Control-2 groups (without surface-water application) for surface application method. Additionally, there was a remarkable difference between the surface roughness values obtained from the two application methods. It is clear that the values of R_a, R_q, and R_z showed a notable decreasing trend with the application of chemicals from the surface. In most cases, these values for the panel groups produced with the surface application method decreased to almost half of those for panels produced with the UF-mixing method. For UF-mixing method, the best results were found in the groups with 5% GA chemical.

It was observed that, especially, surface application of all chemicals provided significant improvements in the surface roughness parameters. This positive effect may be attributed to the increased effectiveness of these chemicals when applied to the surface, resulting in a more uniform surface structure by reducing micro irregularities. Previous studies have reported that parameters in manufacturing and raw material characteristics influence the level of surface roughness (Hiziroglu and Kosonkorn 2006) and, quality of surface for wood and wood-based panels is one of the parameters that has a significant impact on further manufacturing steps (Zhong *et al.* 2013).

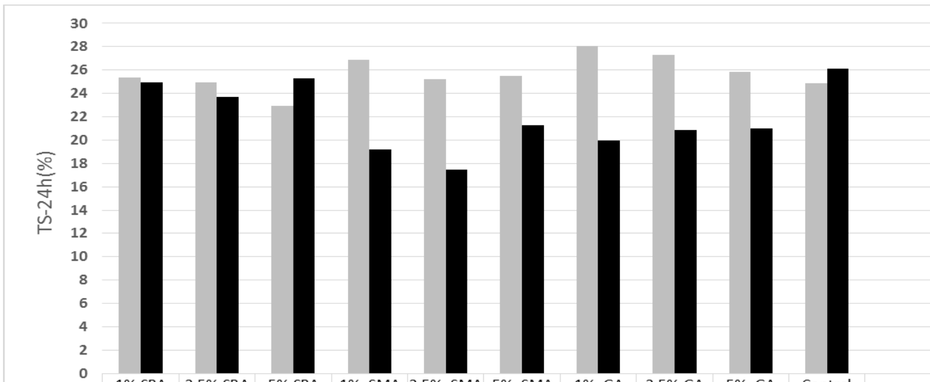
Physical Properties

The thickness swelling and water absorption results are presented in Figs. 4, 5, 6, and 7.



	1%-SBA	2.5%-SBA	5%-SBA	1% -SMA	2.5% -SMA	5% -SMA	1% -GA	2.5%-GA	5% -GA	Control
surface	22,92	22,85	20,68	24,57	23,72	22,32	24,81	24,52	22,45	22,51
UF-mixt	20,42	21,66	22,35	16,03	15,47	17,28	16,76	16,89	17,77	20,54

Fig. 4. TS-2h results of MDF samples

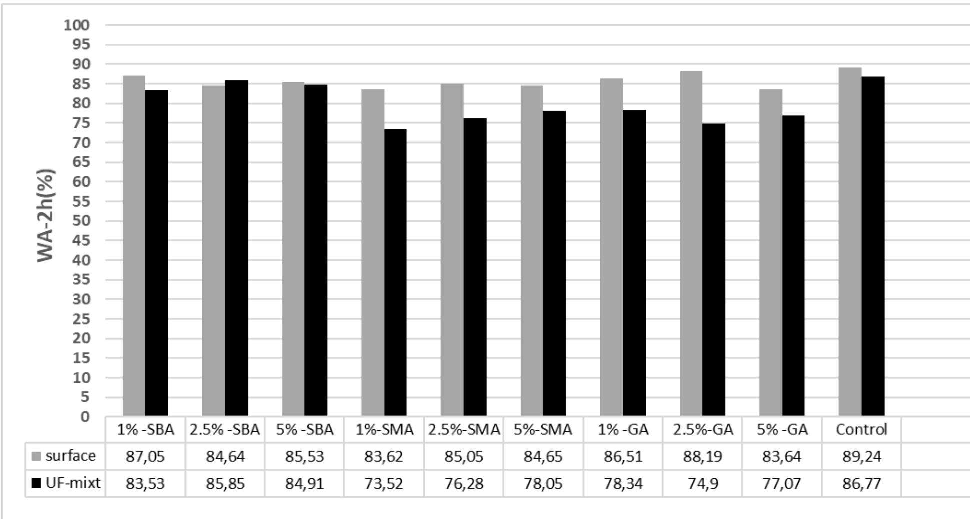


	1%-SBA	2.5%-SBA	5%-SBA	1% -SMA	2.5% -SMA	5% -SMA	1% -GA	2.5%-GA	5% -GA	Control
surface	25,36	24,93	22,97	26,91	25,19	25,53	28,03	27,29	25,86	24,84
UF-mixt	24,95	23,73	25,27	19,18	17,45	21,29	19,98	20,87	21,03	26,14

Fig. 5. TS-24h results of MDF samples

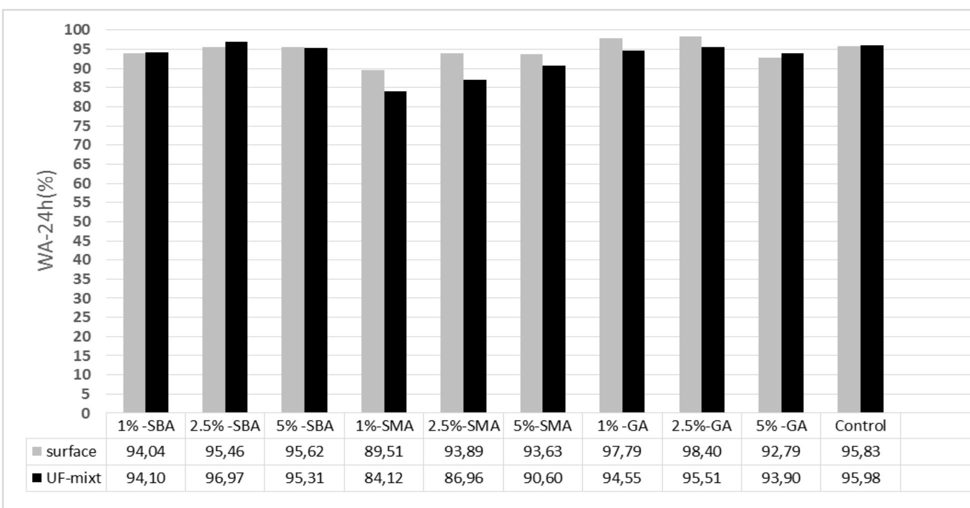
From the data in Figs. 4 and 5, it was observed that the TS values for 2 and 24 h of MDF panels changed depending on the application methods, chemical types and chemical concentrations. TS values (2 and 24 h) ranged from 20.7% to 24.8% and from 23.0% to 28.0% for the groups with the surface application, respectively. These values for 2 and 24 h ranged from 15.5% to 22.4% and 17.4% to 26.1% for the groups with the UF-mixture, respectively. The standard deviations of the 2 and 24 h TS values ranged from 0.21 to 2.10 and 0.43 to 2.20 for the surface application method. These values ranged from 0.67 to 1.52 and 0.31 to 2.01 for the UF-mixing method.

Figures 4 and 5 showed that, generally, only slightly higher or similar TS values (2 and 24 h) were obtained from MDF groups with surface application compared to their control groups. The MDF groups with UF-chemical mixture exhibited lower TS values than their control groups. Additionally, the groups with surface application showed a slightly decreasing trend in TS values with increasing chemical concentrations. Although the UF-mixing method showed unstable trends with small decreases and small increases for the TS values as the chemical concentration increases, these values obtained with this method were noticeably lower than those of the surface application method. The lower values obtained with the UF-mixing method may be due to the homogeneous application of the chemicals mixed into the resin to the fibers before panel production. While the lowest TS values for 2 h and 24 h were obtained with 2.5% SMA for the UF-mixing method, the lowest TS values for 2 h and 24 h were found with 5% SBA for the surface application method. As shown in Figs. 4 and 5, when GA and SMA chemicals were mixed into resin, they showed a better effect on TS values compared to SBA chemical. Mamiński *et al*. (2008) found that while the swelling and water absorption values of particleboards bonded with 5% GA-UF resin decreased, these values increased with the addition of 10% GA.



	1% -SBA	2.5% -SBA	5% -SBA	1%-SMA	2.5%-SMA	5%-SMA	1% -GA	2.5%-GA	5% -GA	Control
surface	87,05	84,64	85,53	83,62	85,05	84,65	86,51	88,19	83,64	89,24
UF-mixt	83,53	85,85	84,91	73,52	76,28	78,05	78,34	74,9	77,07	86,77

Fig. 6. WA-2h results of MDF samples



	1% -SBA	2.5% -SBA	5% -SBA	1%-SMA	2.5%-SMA	5%-SMA	1% -GA	2.5%-GA	5% -GA	Control
surface	94,04	95,46	95,62	89,51	93,89	93,63	97,79	98,40	92,79	95,83
UF-mixt	94,10	96,97	95,31	84,12	86,96	90,60	94,55	95,51	93,90	95,98

Fig. 7. WA-24h results of MDF panels

As can be seen from Figs. 6 and 7, WA (2 h and 24 h) values of MDF panel groups ranged from 83.6% to 89.2% and 89.5% to 98.4% for the groups with surface application, while the WA (2 h and 24 h) values ranged from 73.5% to 86.8% and 84.1% to 97.0% for the groups with the UF-mixture, respectively. For the UF-mixing method, the standard deviations of the WA values (2 h and 24 h) ranged from 1.67 to 2.87 and 1.35 to 2.90, respectively, while the WA values for the surface application method ranged from 4.28 to 9.67 and 3.93 to 9.31, respectively. Similar to the TS values, variable increasing/decreasing trends were observed in WA values as the chemical concentration increased. In particular, no noticeable differences were found among the WA values of the groups produced with the surface application method. For the UF-mixing method, moderate decreases in the WA values (2 h) were determined, only slight decreases were observed in WA (24 h) values. Specifically, for the 24 h WA values, almost similar results were obtained between the chemical-containing groups and the control groups for both methods. The lowest values for 2 h and 24 h were found in the groups containing 1% SMA chemical.

Mechanical Properties

The results of mechanical properties such as internal bond strength (IB), modulus of rupture (MOR), modulus of elasticity (MOE) are presented in Figs. 8, 9, and 10, respectively.

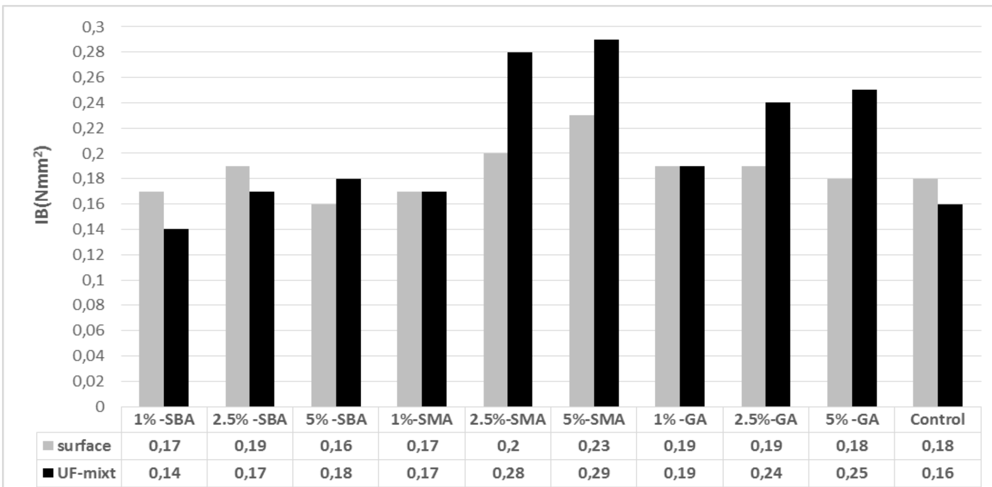


Fig. 8. The IB results of MDF samples

As can be seen in Fig. 8, the IB values of MDF groups showed variability depending on the application methods, chemical types, and chemical concentrations. The IB values ranged from 0.17 N/mm^2 to 0.23 N/mm^2 and 0.14 N/mm^2 to 0.29 N/mm^2 for the surface application method and the UF-mixing method, respectively. The standard deviations ranged from 0.01 to 0.03 and 0.02 to 0.08 for these methods, respectively. While the highest value was obtained for group 5% SMA with the UF-mixture, the lowest value was determined for group 1% SBA with the UF-mixture. Generally, the increasing chemical concentrations resulted in the increasing IB values. The SMA groups showed higher IB values among the other groups.

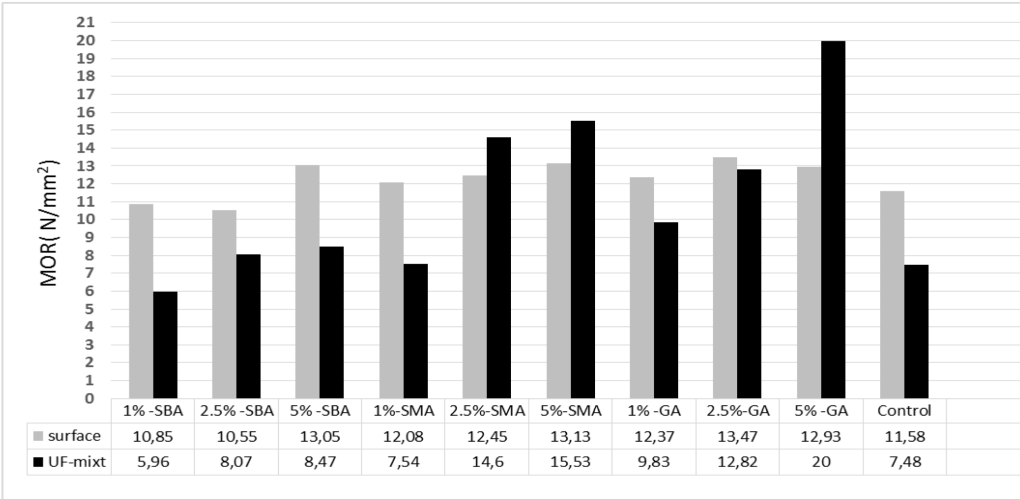


Fig. 9. The MOR results of MDF samples

The surface application method resulted in lower IB values compared to IB values obtained with the UF-mixing method. This may be attributed to the fact that the chemicals used in the surface application method remain only on the MDF panel surfaces. Therefore, these chemicals cannot sufficiently penetrate the interior of the fibers, resulting in a lesser effect on fiber bonding.

Figure 9 demonstrates that the MOR values changed depending on the chemical types, chemical concentrations, and application methods. There have been clear differences between the MOR values of MDF groups produced with two application methods. The values ranged from 10.6 to 13.5 N/mm^2 and 5.96 to 20 N/mm^2 for the groups with surface application and UF-mixing method, respectively. The standard deviations ranged between 0.67 and 1.82 for the surface application method, whereas these values ranged between 0.33 and 2.51 for the UF-mixing method. The MOR values of the MDF groups produced with the surface application method were slightly higher than those of the control group, except for the groups containing 1% and 2.5% SBA chemicals. The chemicals applied to the panel surface did not provide any noteworthy improvement in the MOR values compared to the control group. However, generally higher values were obtained with the UF-mixing method compared to the control group. For UF-mixing method, the 5% GA group showed the highest value, while the 1% SBA group exhibited the lowest value. For surface application method, the 2.5% GA group showed the highest value, while 2.5 % SBA group exhibited the lowest value. Especially, lower MOR values were obtained with SBA chemical among all groups for both application methods.

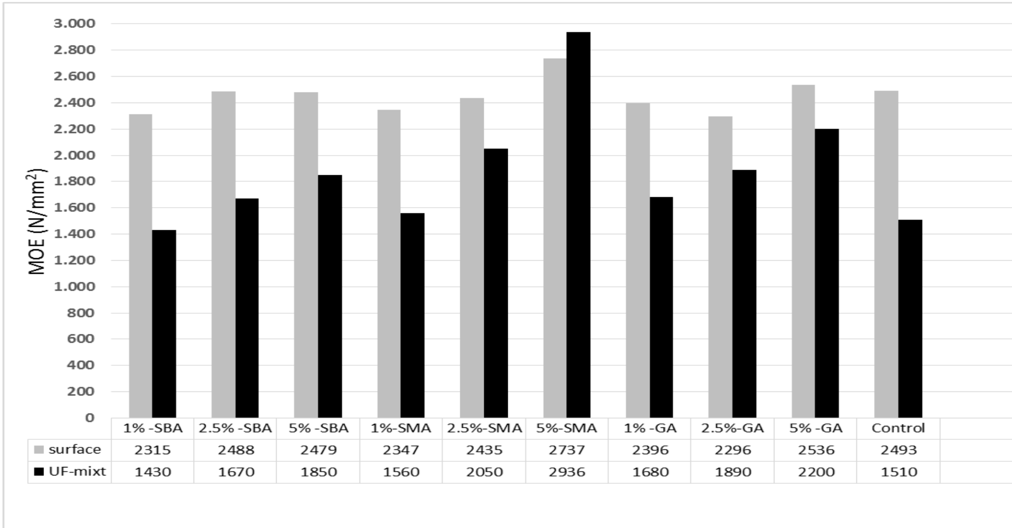


Fig. 10. The MOE results of MDF samples

Figure 10 reveals that the MOE results showed different trend depending on the application methods, chemical types and chemical concentrations. While there were gradual decreases/increases in MOE values of the groups with chemicals compared to control group for the surface application method, clear increases in the MOE values were observed in the groups with chemicals compared to the control group for the UF-mixing method. Generally, the MOE values increased with the increasing chemical concentrations. The MOE values for the groups with surface application and UF-mixing method ranged from 2300 to 2740 N/mm^2 and 1430 to 2940 N/mm^2, respectively. The standard deviations ranged between 96.7 and 390 for the surface application method. These values ranged between 152 and 525 for the UF-mixing method. For the surface application method, while the highest value determined from 5% SMA group, the lowest value was recorded with 2.5% GA group. For the UF-mixing method, while the lowest value was obtained from 1% SBA group, the highest MOE value was obtained with 5% SMA group. When comparing both application methods, it was found that the MOE values obtained from MDF groups produced with UF-mixing method were lower than those obtained from MDF groups produced with the surface application method.

Formaldehyde Emission

The free formaldehyde amount of MDF panel groups depending on application method are given in Fig. 11.

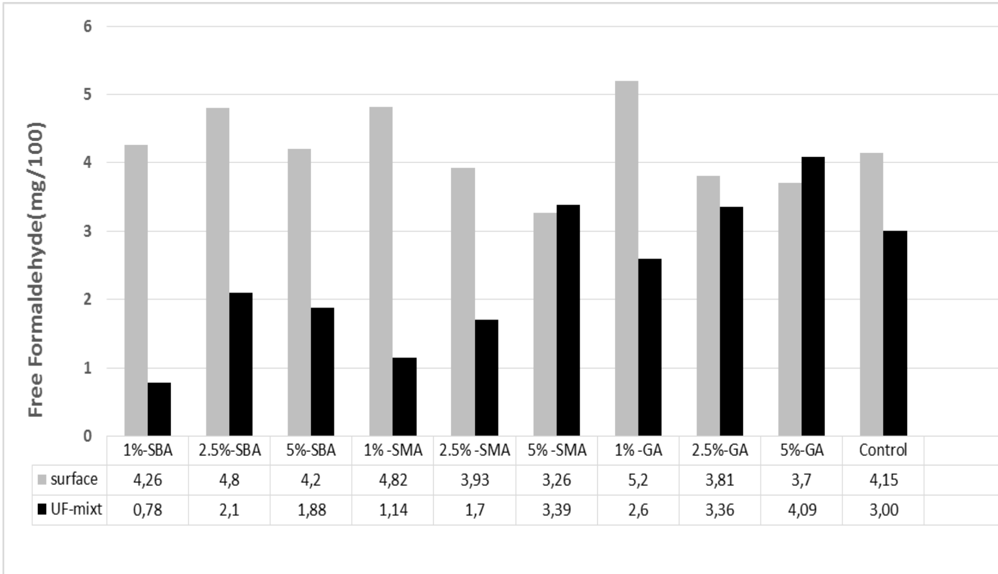


Fig. 11. Free formaldehyde amount of MDF samples

As can be seen from Fig. 11, the free formaldehyde amount of groups changed depending on the application methods, the chemical types, and the chemical concentrations. There were obvious differences among the panel groups. The free formaldehyde amounts ranged from 3.26 to 5.20 mg/100, and 0.78 to 4.09 mg/100 for surface application method and UF-mixing method, respectively. The control values were found to be 4.15 and 3.0 mg/100 for these methods. Generally, remarkably lower values were obtained with panel groups except for 2.5% GA, 5% GA, and 5% SMA groups for the UF-mixing method compared to control group. For this method, the 1% SBA and 5% GA group resulted in the lowest and the highest values, respectively. For the surface application method, while the 1% GA group showed the highest value, the 5% SMA group exhibited the lowest value. When comparing the two methods, better results were obtained with the UF-mixing method. SBA chemical showed markedly lower values than the control and yielded the most favorable results for this method. In contrast, SBA chemical did not yield lower values than the control for surface application method. Although UF-mixing method resulted in lower values, the formaldehyde emission values showed slightly increasing trend with the increasing chemical concentrations. The 1% concentration of all chemicals resulted in the lowest values across all groups for this method.

CONCLUSIONS

The main conclusions of this study are summarized below.

1. The types and concentrations of the chemicals were observed to influence the surface roughness parameters (R_a, R_q, and R_z), depending on the application methods. These parameters showed notable decreases for the surface application method and the urea formaldehyde (UF)-mixing method. The smoother surfaces were obtained in the medium density fiberboard (MDF) groups produced with chemicals for both application methods, compared to the control groups. The surface application method yielded the best results.

2. The thickness swelling (TS) values (2 and 24 h) exhibited different trends for both methods. For the surface application method, slight increasing and decreasing trends were obtained, while for the UF-mixing method, clear differences and a decreasing trends were observed. The UF-mixing method yielded more favorable results. A similar trend was obtained for water absorption (WA) values. UF-mixing method showed a more noticeable decreasing trend in WA values, especially for 2h, compared to the surface application method.
3. The internal bond (IB), modulus of rupture (MOR), and modulus of elasticity (MOE) values generally varied depending on the chemical types, chemical concentrations, and application methods. The surface application method showed lower IB values compared to the UF-mixing method. Generally, the increasing chemical concentrations resulted in the increasing IB values. When comparing the two methods, differences were observed in the MOR and MOE values of the MDF groups. Generally, the MOR and MOE values exhibited similar trends. The MOR and MOE values showed slightly increasing and decreasing trends with the increasing chemical concentrations for the surface application method. However, these values showed only increasing trend with the increasing chemical concentrations for the UF-mixing method.
4. In general, the free formaldehyde levels of MDF panel groups produced using both application methods were reduced. The UF-mixing method yielded the most favorable results compared to the surface application method. The free formaldehyde amounts remarkably decreased for the UF-mixing method compared to the control group. The type and concentration of the chemicals clearly affected the formaldehyde emission levels, depending on the method applied.

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