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Effect of sodium carboxymethyl cellulose (Na-CMC) added to urea-formaldehyde resin on particleboard properties

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Abstract: This study investigated the effect of sodium carboxymethyl cellulose (Na-CMC) added to urea-formaldehyde (UF) resin on particleboard production and its effects on the properties of the boards. The Na-CMC/UF was prepared at the selected blending levels of 0/100, 5/95, 10/90, 15/85, 20/80, 25/75, 30/70, 35/65, 40/60, 45/55, and 50/50. In the production of test panels, 55% coniferous and 45% broadleaved wood chips were used. The physical and mechanical properties of the particleboard panels were determined and evaluated. According to the results, the addition of Na-CMC at the level of between 5% and 20% improved the internal bond (IB) strength by 6%–40%. The best IB strength (0.63 N/mm²) was found in the panels with the 20% Na-CMC admixture. Although an increase in bending strength (modulus of rupture-MOR) was observed with an increase in the amount of Na-CMC used, it was found that the bending modulus of elasticity (MOE) was reduced in comparison with the control sample. The lowest MOE was determined as 1451 N/mm² in the 50% Na-CMC-doped test panels. It was determined that the use of Na-CMC negatively affected the water absorption and thickness swelling values. As a result, it was concluded that UF resin containing up to 20% Na-CMC solution can be used in particleboards produced for applications in which the physical properties are not important.

Key words: Sodium carboxymethyl cellulose, particleboard, urea-formaldehyde resin

1. Introduction

Particleboard is produced from a loose mat consisting of wood material mechanically reduced into small particles such as wood chips, sawdust, sawmill shavings, or other agricultural wastes that are bound together with a synthetic resin or other suitable binders and consolidated under heat and pressure (Istek and Sıradağ, 2013; Mamza et al., 2014; Kevin et al., 2018). Urea-formaldehyde (UF) particleboard is widely applied in furniture manufacturing, interior decoration, acoustical settings, and ceiling and wall coverings. However, UF particleboard is not used in damp or humid environments as its dimensional stability is low (Istek and Sıradağ, 2013; Kevin et al., 2018).

Sodium salt of carboxymethyl cellulose (Na-CMC) is a cellulose ether formed by the reaction of cellulose anhydroglucose unit hydroxyl groups with chloroacetic acid. It exhibits high biocompatibility and biodegradability and low immunogenicity and is easily dissolved in water. It is used as a viscosity modifier, thickener, emulsion stabilizer, and water retention agent in food, cosmetics and paints (Colombo et al., 2000; Ugwoke et al., 2000; Kono, 2014). It has been emphasized that the binder produced by

adding 7.5% carboxymethyl cellulose (CMC) solution into sodium silicate exhibits optimal levels for adhesion and water resistance compared to other treatments (Zhang et al., 2011). Urea formaldehyde (UF) resin is widely used in particleboard production because it is inexpensive, easy to use, and possesses sufficient strength. The formaldehyde/urea molar ratios in the production of the resin determine its adhesion efficiency. As the molar ratio of formaldehyde in the solution increases, the adhesion efficiency increases. However, more free formaldehyde is released during and after hardening, adversely affecting the environment and human health (Rocket, 1997; Eroğlu and Usta, 2000; Zhang et al., 2011; Hematabadi et al., 2012; Park and Causin, 2013; İstek et al., 2018). It has been noted that CMC can be used for the modification of viscosity, solids content, and the binding performance of starch and isocyanate-like binders, and for improving the compatibility between binders and prepolymerization (Qiao et al., 2014). The properties and formaldehyde emission of panels are affected by Na-CMC-doped UF resin, but this effect is not linear. Furthermore, it has been emphasized that the average modulus of elasticity (MOE) of panels containing 10% Na-CMC increased by

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6.65%, and with 20%, and 30% Na-CMC supplementation, the free formaldehyde emission was significantly reduced, while all other properties were adversely affected (Özlüsoylu and İstek, 2018). It was observed that the bending strength and the tensile strength perpendicular to the surface of the composites obtained by using CMC copper sulphate binder and sugarcane pulp were higher than in the composites produced with UF resin. It was pointed out that this was due to the strong bonds formed between the Cu (II) ions and the sugarcane fibers (Basta et al., 2004).

The purpose of this study was to investigate binders prepared with specific ratios of Na-CMC additive to UF resin for use in particleboard production, including determination of the effect of the Na-CMC additive ratios to UF resin on the physical and mechanical properties of test panels and on the formaldehyde emissions of the binders.

2. Materials and methods

2.1. Material

The wood chips used in this study were readily obtained from a commercial particleboard operation. The chips, taken from the core and face layers, were 55% from coniferous and 45% from broadleaved species in origin. The chips were oven-dried at 120 ± 5 °C to 1%–3% humidity. Sodium carboxymethyl cellulose (Na-CMC), known under the trade name DENCELL T-30, was used as an adhesive additive to the UF resin composed of 50% solids with 1% ammonium chloride (NH_4Cl) solution hardener based on the resin solids. The properties of the Na-CMC used as an additive to the UF resin are given in Table 1.

2.2. Method

2.2.1. Preparation of Na-CMC/ UF solutions

The hybrid UF resin used in the production of the test panels consisted of a mixture of 50% UF and 7% Na-CMC solutions. The amounts of Na-CMC mixed with the UF were selected according to the dry substance content of the resin and were added as 0% (Control), 5% (A), 10%

(B), 15% (C), 20% (D), 25% (E), 30% (F), 35% (G), 40% (H), 45% (I), and 50% (J). The amounts used were 10% in the surface layers and 8% in the middle layer, according to the full dry-chip weight. In addition, a solution of 20% ammonium chloride (NH_4Cl) was added to the resin solution at a level of 1% as a hardener.

To prepare the 7% (w/w) aqueous solution of Na-CMC, 7.7 g of granular Na-CMC were mixed with 92.3 mL of water (20 °C) using a mechanical stirrer at speeds of 900–1700 rpm for 20 min. The viscosity of the obtained Na-CMC solution was measured as 150–200 cP. This value was selected because of its similarity to the viscosity of the UF resin. The UF and Na-CMC solutions at the abovementioned ratios were prepared by depositing them in a beaker and mixing at a speed of 1000 rpm for 5 min.

2.2.2. Test panel manufacture

In the production of the test panels, the hybrid UF resin having a solid content of 50% was used, according to the full dry particle weight, as 10% in the surface layers and 8% in the middle layer and was applied using a spray gun with a nozzle diameter of 1.7 mm at a rotational speed of 50 rpm and a pressure of 6 bar. The panels were formed using a wooden mold with the dimensions of $400 \times 400 \times 300$ mm. The spreading process was performed manually so that the middle layer chip rate was 60% and the top and bottom layer chip rate was 40%. The test panels were produced in 3 layers with a 16-mm thickness and 680 kg/m³ density under conditions of 175 ± 5 °C for 5 min at a pressure of 160–170 bar.

2.2.3. Preparation of test samples and evaluations

Test specimens and their dimensions were prepared in accordance with TS EN 325 (2012) and TS EN 326-1 (1999). After cutting, each test sample was conditioned at 20 ± 2 °C and $65 \pm 5\%$ RH for 2 weeks before testing. Physical and mechanical properties of the test samples were determined according to relevant standards. Specific gravity ($n = 8$), water absorption (WA; $n = 8$) and thickness swelling (TS; $n = 8$), internal bond strength (tensile strength perpendicular to the surface of the board) (IB; n

Table 1. Properties of Na-CMC (DENCELL T-30).

Properties	Standard	Measurement	Methods
Moisture content (%)	max. 7.0	6.0	IS 3220-1966
Active substance (%)	min. 55.0	56.5	IS 3220-1966
Substitution level	0.85 – 1.00	0.85	IS 3220-1966
Dry matter (%)	Max. 45.0	43.5	IS 3220-1966
pH (25 °C 1% Solution)	8.0 – 10.0	9.4	WTW inoLab pH Level 1
Viscosity (2% solution)	450 - 650	570	Brookfield Lvdv-I

= 6), bending strength (modulus of rupture) (MOR; n = 6), and modulus of elasticity in bending (MOE; n = 6) were determined using TS EN 323 (1999), TS EN 317 (1999), TS EN 319 (1999), and TS EN 310 (1999), respectively. Formaldehyde emission was determined according to the TS 4894 EN 120 (1999) perforator method. The obtained data were statistically evaluated by the SPSS statistical software program, and the effects of the ratio of Na-CMC added to the UF resin on the properties of the particleboard were determined. One-way analysis of variance (ANOVA) was performed ($P < 0.05$) in order to determine statistically significant differences among the factors. In addition, results were analysed for meaningful differences among the groups using the Duncan test.

3. Results and discussion

The results of the test panels for average density, water absorption, and thickness swelling (2 h and 24 h), standard deviation values and differences between the groups are given in Table 2.

Table 2 shows that there were no significant differences between the densities. When the WA and TS data were examined, it was found that there were statistically significant differences among the test panel groups. With the increase of the Na-CMC contribution level to the UF resin, the WA and TS levels increased, and this increase was higher in the G, H, I, and J groups. According to these results, it was understood that as the ratio of Na-CMC usage increased in the UF resin modification, the WA and TS properties of the panels were affected negatively. However, these values were slightly lower up to the Na-

CMC doping levels of 15% for WA and 20% for TS, compared to the control samples, although this decrease was not statistically significant ($P < 0.05$).

Data for the mechanical properties of IB strength, MOR, and flexural MOE are shown in Table 3. When the table is examined, it can be seen that the mechanical properties changed according to the doping level of Na-CMC in the UF resin solution. In the statistical analysis, it was calculated that these changes were significant ($P < 0.05$) among many groups, whereas they were not significant among others.

The changes in the IB strength are seen in Figure 1. In the IB strength tests, it was observed that the break usually occurred in the core layer of the panels and that up to the doping level of 30% Na-CMC, the IB strength increased, after which it decreased. According to the ANOVA analysis, there were significant differences among the test panel groups at a 95% confidence level due to the Na-CMC addition levels. The groups of tested panels in which IB strength was statistically significant were determined by the Duncan test. The Duncan test results are shown in Table 3 and include numerical data and graphs with letters indicating significance levels. The 20% Na-CMC-doped D group was found to have the highest increase in IB values. This increase was calculated according to the 40% average of the control test panels. As Na-CMC is produced from alpha cellulose, it has the potential to increase IB strength as it can easily bind with the wood chips. It is stated that CMC has high molecular weight and thickening ability and contributes to adhesion strength by improving interface properties (Cao et al. 2001). Similarly, Qiao et

Table 2. Physical properties of tests samples.

Test Panel Groups	Density (g/cm ³)	Water absorption (WA) (%)		Thickness swelling (TS) (%)	
		2 h	24 h	2 h	24 h
Control	0.68 ± 0.01 a	87.68 ± 3.52 ab	103.37 ± 4.06 a	27.71 ± 2.94 a	31.11 ± 3.33 a
A	0.68 ± 0.01 a	86.49 ± 6.72 a	103.92 ± 8.09 a	29.41 ± 4.98 a	33.14 ± 5.85 ab
B	0.69 ± 0.01 a	84.46 ± 6.01 a	104.20 ± 9.23 a	30.20 ± 3.64 ab	33.39 ± 4.81 ab
C	0.68 ± 0.01 a	93.39 ± 7.88 bc	108.56 ± 7.63 ab	31.00 ± 3.17 ab	35.15 ± 3.83 ab
D	0.69 ± 0.01 a	95.76 ± 5.49 c	111.72 ± 6.98 bc	31.69 ± 3.53 ab	35.30 ± 3.97 ab
E	0.69 ± 0.01 a	98.85 ± 6.75 c	111.94 ± 6.26 bc	34.85 ± 4.95 bc	38.46 ± 5.09 bc
F	0.69 ± 0.01 a	100.03 ± 7.08 c	115.86 ± 8.17 c	37.45 ± 5.19 c	42.36 ± 6.50 c
G	0.67 ± 0.02 a	110.47 ± 10.38 d	126.02 ± 11.09 d	41.88 ± 4.72 d	50.81 ± 6.86 d
H	0.68 ± 0.01 a	122.38 ± 8.16 e	134.81 ± 5.38 e	51.54 ± 6.19 e	65.56 ± 8.50 e
I	0.67 ± 0.01 a	127.81 ± 5.57 ef	135.38 ± 5.52 e	55.33 ± 7.02 e	65.77 ± 5.00 e
J	0.67 ± 0.02 a	129.26 ± 6.09 f	134.10 ± 6.76 e	55.90 ± 5.63 e	63.47 ± 3.54 e

±: standard deviation. Means followed by the same letters (a,b,c,d,e,f) in the same column are not significantly different ($P < 0.05$).

Table 3. Mechanical properties of test samples.

Groups	IB (N/mm ²)	MOR (N/mm ²)	MOE (N/mm ²)
Control	0.45 ± 0.03 de	16.26 ± 0.41 d	2921 ± 52 g
A	0.48 ± 0.05 ef	16.34 ± 0.96 d	2722 ± 130 f
B	0.53 ± 0.03 gh	18.27 ± 0.62 e	2602 ± 53 f
C	0.57 ± 0.06 i	14.61 ± 0.58 c	2605 ± 106 f
D	0.63 ± 0.06 j	13.71 ± 0.73 bc	2331 ± 94 e
E	0.56 ± 0.035 hi	13.06 ± 0.95 b	2107 ± 105 d
F	0.50 ± 0.05 fg	11.71 ± 0.73 ab	1933 ± 124 c
G	0.42 ± 0.06 d	10.77 ± 0.66 a	1900 ± 114 c
H	0.38 ± 0.04 c	10.77 ± 0.34 a	1750 ± 121 b
I	0.33 ± 0.04 b	10.28 ± 0.36 a	1619 ± 146 b
J	0.28 ± 0.03 a	10.33 ± 0.46 a	1452 ± 167 a

±: standard deviation. Means followed by the same letters (a,b,c,d,e,f,g,h,i,j) in the same column are not significantly different (P < 0.05).

al (2014) stated that the adhesion property first increased and then decreased with increasing CMC concentration. On the other hand, the IB strength of the 50% Na-CMC-added J group panels decreased by 37.7%. CMC forms a very weak bond compared to UF. It is thought that IB strength also decreases as the UF concentration decreases in total glue after the addition of 20% Na-CMC. Up to the addition level of 35% Na-CMC, all test panel groups were able to meet the requirements of particleboard Type P2 for use in indoor applications (including furniture) (TS EN 312, 2012). Özlüsoylu and İstek (2018) emphasized that in particleboard production, the IB increased slightly and then decreased depending on the addition level of Na-CMC to the UF resin.

Figure 2 shows the effect of the addition of Na-CMC to the UF resin used in particleboard production on the MOR. The Na-CMC additions to the UF resin are seen to increase and then decrease the MOR value. There was no statistically significant difference between the control and Group A test panels. The highest MOR value was found to be 18.27 N/mm² in the B group and the lowest 10.28 N/mm² in the I group. Compared to the control sample, the maximum MOR value of group B was higher by 12.36%, while it was 36.78% lower in the I group. The MOR value increased up to the addition level of 10% Na-CMC to the UF resin. However, the IB value increased up to the level of 30% Na-CMC addition compared to the controls. This was thought to have been caused by the Na-CMC forming a weaker bond than that of the UF resin. It is stated in the TS EN 312 (2012) standard that the MOR strength for Type P2 particleboard should be greater than 13 N/mm². Therefore, Na-CMC can be added to UF resin up to a level of 25%.

The relation between the Na-CMC addition ratio to the UF resin used in particleboard production and the MOE is shown in Figure 3. According to the control test sample, the Na-CMC contribution to the UF resin adversely affected the MOE data. This effect was found to be statistically significant among many groups at a 95% confidence level. It was determined that the MOE feature decreased linearly, and the mean in the lowest group (J) was 1452/mm². Compared to the control sample, the reduction was calculated to be up to 50%. However, according to the TS EN 312 standard, 35% Na-CMC can be added to UF resin for Type P2 particleboard. The decrease in MOR and MOE with increasing CMC concentration can be attributed to the hydrophilic structure of CMC.

Figure 4 shows the effect of the addition of Na-CMC to UF resin used in particleboard production on formaldehyde emission.

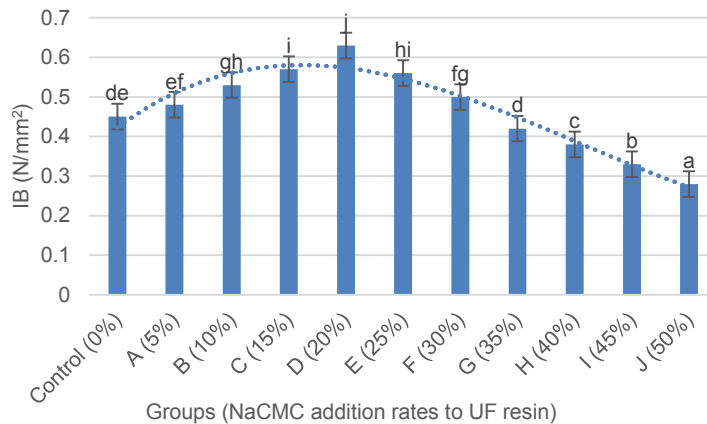


Figure 1. Correlation between IB strength and addition levels of Na-CMC to the UF resin.

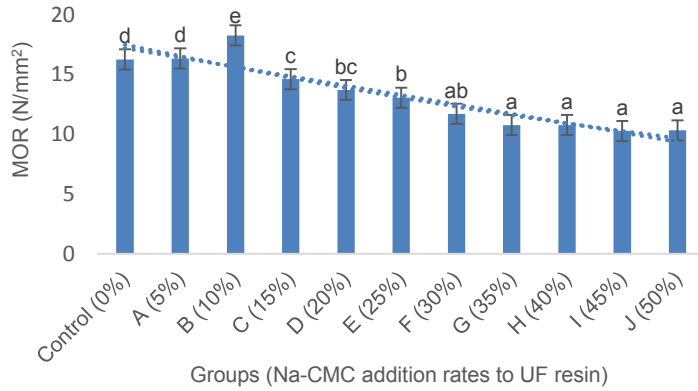


Figure 2. Correlation between MOR and addition levels of Na-CMC to the UF resin.

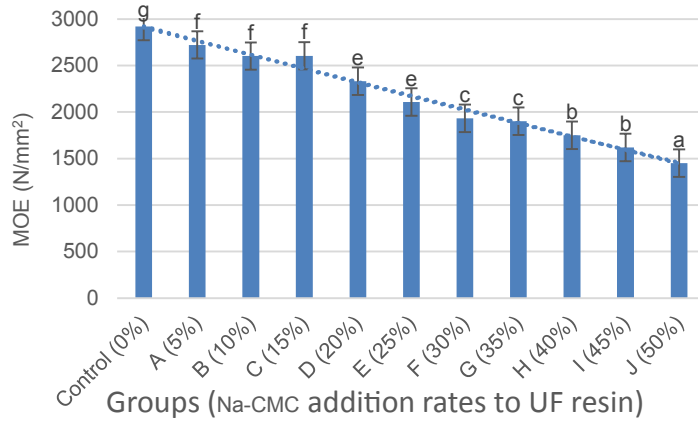


Figure 3. Correlation between MOE and addition levels of Na-CMC to the UF resin.

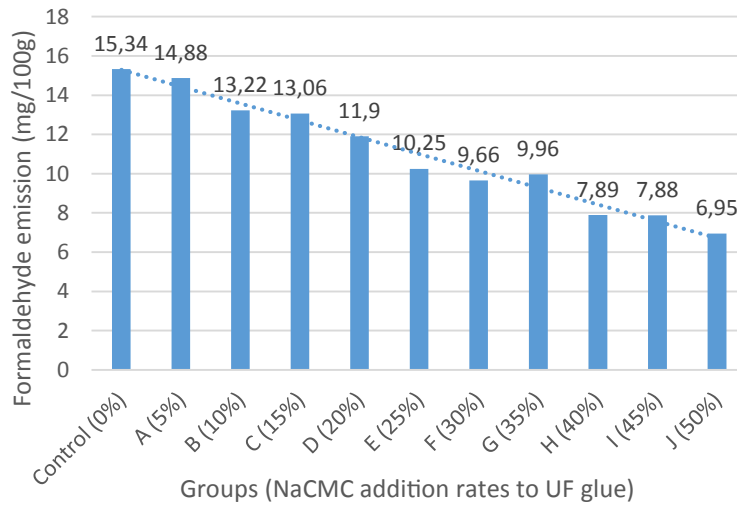


Figure 4. Correlation between formaldehyde emission and addition levels of Na-CMC to the UF resin.

When Figure 4 is examined, a linear decrease in the amount of free formaldehyde emission is observed with the increase in the level of Na-CMC added to the UF resin. The lowest formaldehyde emission was detected in J group panels containing 6.95 mg/100 g and 50% Na-CMC, and the highest formaldehyde emission was found in the control group without Na-CMC with 15.34 mg/100 g. Depending on the amount of Na-CMC usage, when compared with the control group, the formaldehyde emission value was calculated to decrease by 2.99%-54.69%. It was understood that the emission of the panels with Na-CMC usage levels of 40% and above would meet E1 class specifications. As the total amount of resin used as a binder in the production of the test panels remained constant, the amount of UF decreased in relation to the amount of Na-CMC added; thus, the amount of formaldehyde emission was expected to decrease. However, it has been reported that the use of modified starch has an effect on the formaldehyde emission of UF resin and that the reaction between carboxyl and methylol urea reduces emission (Zhu et al., 2014).

4. Conclusions

The adhesive used in the production of wood composite panels is an important component in particleboard production costs. UF resin is widely used because it enables the inexpensive production of a board of the desired quality. Thus, in this study, the aim was to reduce resin costs by adding Na-CMC solution to the UF resin

in particleboard production, and produce panels of the desired quality at lower cost. Therefore, the study sought to determine the changes in the properties of the board depending on the level of Na-CMC addition. In light of the findings, the following conclusions were reached:

- The thickness swelling and water absorption properties of the test panels decreased with the increasing levels of the Na-CMC additive. However, the water absorption at 15% and thickness swelling at 20% Na-CMC admixture levels were not statistically significant compared to the control samples.

- The IB strength increased up to the level of 30% Na-CMC addition and then decreased thereafter. In applications where bending resistance is important, mixtures of up to 25% Na-CMC can be used in UF resin.

- UF resin with Na-CMC addition levels of up to 35% meets the standards for MOE and IB properties and can be used for Type P2 particleboard.

- As the amount of Na-CMC additive to the UF resin increased, the physical properties of the particleboard decreased. On the other hand, the mechanical properties of MOR and IB strength initially increased and then decreased. The MOE was found to decrease linearly as the addition level increased.

- As the Na-CMC substitution levels can be important, studies should be carried out to investigate Na-CMC solutions in relation to different properties.

- It was calculated that Na-CMC has at least 50% lower cost than UF.

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