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Effects of Activated Carbon on Medium Density Fiber Board Properties

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Abstract: The negative effects of formaldehyde emission in MDF production on human health are known, and therefore many scientists are working to reduce formaldehyde emissions. In this study, the effects of MDF plates on formaldehyde emission were investigated by adding different amounts of activated carbon into urea formaldehyde resin during the synthesis phase. First, the gelation time behavior of the resin was studied by observing the pH, gelation time, solid content, flow time and viscosity of the modified resin in comparison to the standard reference resin which has no activated carbon inside. The dosing of the activated charcoal in the dry resin was kept at 1wt%, 3wt% and 5wt%. After that modified resin was used in the production of 40x40 cm^2 MDF samples by using laboratory scale press line with full automation system. İnternal bonding strength, surface soundness, screw holding resistance, water absorption and thickness swelling were also measured in addition to the main interested parameter formaldehyde emission level which is determined via spectrometric technique following an extraction procedure. Threshold values for activated carbon were determined to be 1wt%. Formaldehyde emission level was observed where addition of 1wt% activated carbon into the urea formaldehyde adhesive decreased the formaldehyde emission 52% comparison to reference whereas addition of activated carbon at above its threshold level provided 47% decreasing.

Keywords: activated carbon, formaldehyde emission, MDF, adsorbent.

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1. INTRODUCTION

The increase in diseases in the world has increased the awareness of the chemical hazards that will come from the products used. For this reason, it has become an important issue to reduce the chemicals released over time from the products that are constantly used in furniture industry. Medium density fiber boards (MDF) that are used different areas school, home, etc. is an important wood panel industry composite material consisting urea formaldehyde. Urea formaldehyde resin has an important use in wood panel industry. This resin is preferred because it is cheap and transparent but ıt has disadvantages. These disadvantages are low water resistance and high formaldehyde emission that is measured from MDF. As it known, formaldehyde has many negative effects on human health. One important among these is the increasing risk of cancer. In 2004 the Internal Agency for Research on Cancer (IARC) classified formaldehyde as harmful chemical for human body (Pizzi 1994; Beram and Yasar, 2020).

Formaldehyde is used in the production of urea formaldehyde resins used in the MDF industry and depending on the reaction conditions between urea and formaldehyde, some amount formaldehyde can remain in the environment without reacting. In addition, some formaldehyde is released due to bond formation in the condensation stage of the resin, which develops during the pressing stage of the MDF production. Due to these reasons, some formaldehyde which is called free formaldehyde remains in the fiber board plate produced (Pizzi 1989).

Consequently, the use of formaldehyde on wood panels currently has been reduced to particular levels, regarded as not harmful to human health. Formaldehyde emission can be lowered by several methods. Several methods for producing low formaldehyde emission MDF panels have been studied, such as reducing formaldehyde to urea mol ratio and addition of formaldehyde scavengers in to resin. However mostly mechanical and physical properties of wood based panels have been affected badly. In addition to this, decreasing formaldehyde mol ratio causes to spread of curing time at MDF production. In this situation requires more energy and time. In the 21st century, where energy and time are important, this is an undesirable situation. The other factors affecting formaldehyde emission are wood type, resin type, type of hardeners, press conditions, amount of resin used in MDF production and storage time. Moreover, modifications of the resin with different amine containing chemicals are also important to reduce formaldehyde emission.

Activated carbons have been used as adsorbents in various fields, for instance, solvent recovery, gas separation, and deodorization. The activated carbon is characterized by a strong adsorption capacity which is attributed to its large internal surface area, porosity and high degree of surface reactivity. In related to this the use of activated carbon is one of the possible methods to reduce formaldehyde emission (Kumar et al., 2013).

The use of activated carbon as formaldehyde absorbent has been analyzed by many researchers rayon based activated carbon as formaldehyde absorbent and activated charcoal have been used as bio-scavenger for decreasing formaldehyde emission from melamine formaldehyde resin.

As the relevance, in this work aimed to investigate the effect of activated carbon addition in to urea formaldehyde resin properties, formaldehyde emission values of MDF, mechanical and physical properties of MDF.

2. MATERIALS AND METHODS

2.1. Materials

Urea and formaldehyde to be used in the resin synthesis was provided from AGT AĞAÇ SAN.TİC.A.Ş. Mixed wood fibers which contain of soft and hardwood fibers that were be used in the MDF productions were provided by AGT. The activated carbon powders that have 200 mesh particle size, 900-950 m^2/g surface area, iodine number greater than 900 and pH 8-10 were procured from ECS KİMYA.

2.2. Synthesis of urea formaldehyde resin

Urea formaldehyde resin synthesis basically; it is dived into two stages: an alkaline condensation stage in which mono-, di- and trimethylolureas forms are formed, and a condensation stage of the formed methylolureas in acid environment. Table 1 shows the basic steps of the resin manufacturing parameters.

In the synthesis process, 45% industrial type aqueous formaldehyde solution and powdered urea have been used. The mol ration of formaldehyde/urea was taken as 1.04/1.00. Pure water was added by weighing powder urea in the appropriate mole ratio into three-necked glass reaction ballon flask assembly, then placed in heated magnetic stirrer unit and set to heat at 40 ºC. At this stage, the appropriate molar ratio of formaldehyde was added gradually and the pH of the reaction media was adjusted to 8.20 with 20% NaOH solution by weight. Reaction was continued at 40°C for 30 minutes. Then, for polycondensation, the pH was arranged with ~4.5 with formic acid. The reaction was continued at 90 ºC for 100 – 120 minutes by controlling flow time of resin with DIN Cup 4. Finally, while the resin was cooled to 70 ºC, its pH was adjusted to 8.5 and the reaction was continued for a while. Finally, vacuum drying was applied to the solution and resin was cooled to 40 °C, and the solid content of resin was reduced to 58% from %60 by weight.

In order for the synthesized 1.04 mol urea formaldehyde resin to cure sufficiently in the plate press stage, it must be used with a hardener. As the hardener 20% by weight aqueous ammonium chloride was used, constituting 4% by weight based on the resin solid content.

Table 1. Resin manufacturing parameters

Resin manufacturing parameters	
Parameters	**Values**
pH	8.20 ±10
Viscosity (cP@ 25°C)	160 cP±10 at 30 rpm
Flow time (second @ 25°C)	25±5
Gelation time (second)	60±5
Solid content (%)	58±1

2.3. Mixing of activated carbon with urea formaldehyde resin

To obtain a uniform dispersion of activated carbon powder in the urea formaldehyde resin, mechanical stirring with YOKEŞ VBR-600 high shear disperser mixer was done for 30 min at 1200 rpm by using cowls type blade. Activated carbon was added to the urea formaldehyde resin at 1%, 3% and 5% by weight according to the resin solid weight. The modified resin was named based on percent added as AC1, AC3 and AC5. AC0 indicates reference resin. AC0 shows that absence of activated carbon powder in the resin.

2.4. Characterization of physical properties of activated carbon containing urea formaldehyde resins

Viscosity measurements were done by Brookfield LV DV2T viscometer by using spindle no 1 at 30 rpm 25°C. Flow time measurements were done by DIN cup 4mm. Gelation time tests were done by using water bath at 100 °C with stirring according to related standard test method.

2.5. Preparation of medium density fiberboard and physical and mechanical testing

Table 2 shows the basic production parameters of MDF boards containing different amounts of activated carbon. The resin free wood fibers (a mixture of 15% beech wood fiber + 85% pine wood fiber) with an average moisture content of 30% were dried in an ındustrial oven for approximately 6 hours until 2% - 4% humidity was achieved. Theoretically the amount of dry fiber was calculated and activated carbon added urea formaldehyde resin was weighed as 12% according to dry fiber amount. Activated carbon added resin was sprayed onto the wood fibers with the help of a mixer with a nozzle system, and a homogenous glue fiber mixture was tried to be obtained. After a 3 g of fiber sample taken from the resinated wood fiber mixture and analyzed in a moisture analyzer and, it was determined that it had an average moisture content of 9% - 10%, and this value was appropriate for pressing. The glued fibers that activated carbon added resin were transferred into a 40x40 cm^2 mold with the help of a vacuum suction unit and the preform was formed before the press. Then, it was transferred to the IMAL PAL laboratory press unit and pressed with a pressure of 120 N/cm^2 for 326 seconds. Table 3 shows all the details of the MDF that contains activated carbon added resin and for reference MDF. The boards were then conditioned to attain uniform moisture content in panels. After that, the boards were cutted and tested according to related standard test method for determining of internal bond strength (EN 319), Edge screw holding resistance (EN 320), and surface soundness (EN 311). Physical tests of samples as thickness swelling and water absorption (EN 317), moisture content determination were done (EN 322). The mechanical properties of MDF panels were evaluated according to TS EN 622-5. Internal bonding tests and other mechanical tests were done with universal testing machine (IMAL IB800 Board property tester)

Table 2. MDF manufacturing parameters with different loading activated carbon

MDF manufacturing parameters	
Parameters	**Values**
Size	400*400 mm
Thickness	17 mm ± 1
Target density	740±20 kg/m^3
Press Pressure	120 N/cm^2
Pressing Time	326 seconds
Press temperature (for both top and bottom plate)	190 °C
UF resin wt % of dry wood fibers	12wt%
Activated carbon wt % of solid resin content	1%, 3% and 5%
Number of boards for each type of concentrations	4

2.6. Formaldehyde emission testing

The formaldehyde emissions from MDF panels were evaluated using the EN-120 (perforator method). 100 g sample were put in a round bottomed flask that contain the 600 mL of toluene. The 1000 mL of distilled water was poured into the perforator attachment. The samples were boiled with the toluene for 2 hours. In this test method the distilled water absorbs the formaldehyde and the volatile organic compounds captured by the boiling toluene. Formaldehyde trapped by the water is then quantitatively determined using UV spectrophotometer.

3. Results

3.1. Effect of activated carbon on the resin physical properties

As shown in the table 3, increasing with amount of activated carbon increases the viscosity of urea formaldehyde resin and extended the gelation time period.

Table 3. Resin properties with addition activated carbon

Sample	pH	Flow Time (second @ 25°C)	Gelation Time (second)	Viscosity (cP@ 25°C)	Solid Content (%)
Reference	8.15	20.00	53	164	58,76
AC1	8.30	20.12	66	174	59,12
AC3	8.52	23.91	88	197	59,69
AC5	8.63	25.13	96	227	60,15

The reactivity of the UF resin depends on the amount of free formaldehyde which produces more acidic during the curing process when the hardener is added (Moslemi 2020). The pH values in Table 3 are the values measured only with the activated carbon, without the addition of hardener. Gelation time test were done with adding the hardener ammonium chloride solution. Since the pH of the resin medium is high, we expect the gel time to be extended. This situation was parallel to the literature. The high pH value of the activated carbon increased the pH value of the resin and extended the gel time period even after the addition of hardener, since the environment was not acidic enough with the increasing concentration of activated carbon.

Resin flow time, viscosity and solid content increased with the addition of activated carbon. Increasing the resin viscosity and flow time will decrease the resin fluidity and cause a decrease in the adhesive property. This situation may cause weakening of the mechanical strength of MDF (Anjum 2020).

3.2. Physical and mechanical properties of MDF panels

Physical tests were done as water absorption and thickness swelling for 24h. For the water absorption tests, the test results for the MDF samples coded as AC0, AC1, AC3 and AC5 are respectively; it is 48.13%, 49.19%, 45.92% and 53.1%. The results are shown that in figure1.

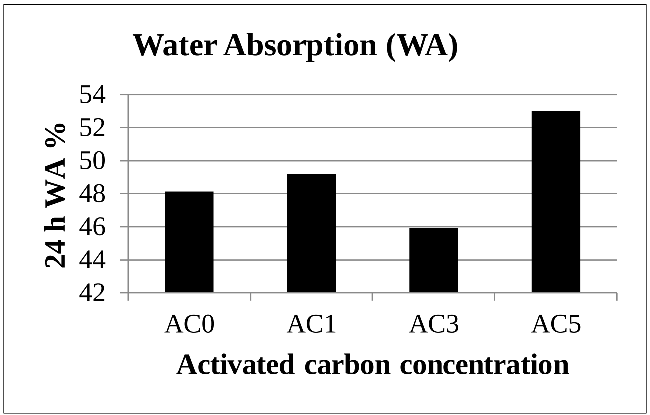

Figure 1. Water Absorption results of MDF panels

For the thickness swelling tests, the results for the MDF samples coded as AC0, AC1, AC3 and AC5 are respectively; as shown in figure 2, it is 19.48, 19.66%, 19.46% and 22.01%.

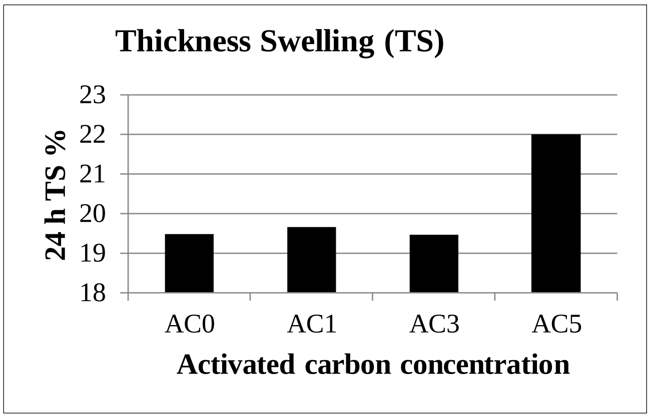

Figure 2. Thickness swelling results of MDF panel

When the results of MDF samples were evaluated, there was an increase of 0.91% in swelling value compared to the reference at 24 hours swelling tests with the addition of 1% activated carbon. There was a 10% decrease and 13% increase for the 3% and 5% concentrations respectively. Based on the results, it was observed that the addition of 1% activated carbon did not cause a significant increase in swelling value. Addition of 3% active carbon caused a decrease in swelling value. When all concentrations are evaluated ıt can be stated that the threshold value is 3% for the swelling test because the addition of 5% activated carbon negatively affected the swelling value of the system within increase of 13%. With the addition of activated carbon, the change in water absorption values at increasing values at increasing rates compared to the reference is an increase of 2.2%, a decrease of 4.6% and an increase of 10.4% respectively.

Table 4. Thickness swelling and water absorption values of MDF panels

Sample	Density kg/m^3	Moisture %	24 h TS %	24 h WA %
Reference	756.64	4.93	19.48	48.13
AC1	753.48	4.99	19.66	49.19
AC3	758.37	4.63	19.46	45.92
AC5	755.34	4.90	22.01	53.01

According to table 4 thickness swelling (TS) and water absorption (WA), activated carbon particle addition did not affect the moisture and density values of the MDF panels, and did not cause a significant change in TS and WA values. This was also observed in the study of in a literature (Kumar et al, 2013).

Mechanical tests were done as internal bonding resistance, surface soundness and screw holding resistance. For the internal bonding tests, the results for MDF samples coded as AC0, AC1, AC3 and AC5 are respectively it is 0.32 N/mm2, 0.34 N/mm2, 0.32 N/mm2 and 0.30 N/mm2. The results are shown in figure 3. The results for surface soundness tests are respectively 0.80 N/mm^2, 0.69 N/mm^2, 0.75 N/mm^2, and 0.84 N/mm^2. The results are shown in figure 4. For the screw holding resistance tests, the results for MDF samples coded as AC0, AC1, AC3 and AC5 are respectively it is 690.00 N, 737.50 N, 668.50 N and 660.75 N. The results are shown in figure 5.

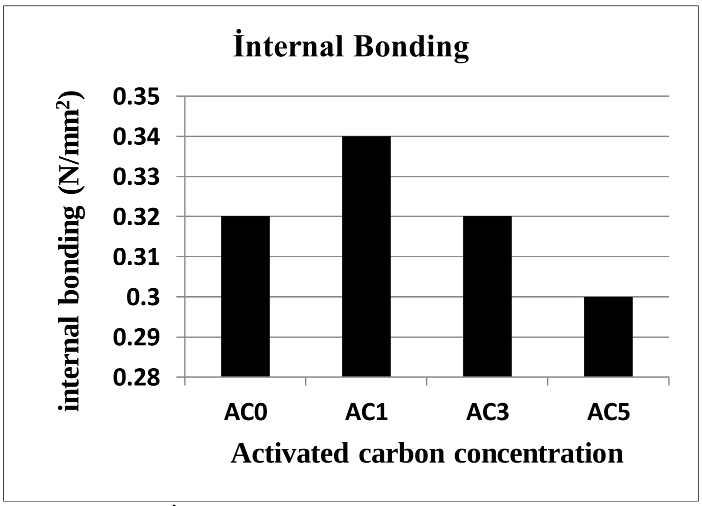

Figure 3. İnternal bonding results of MDF panels

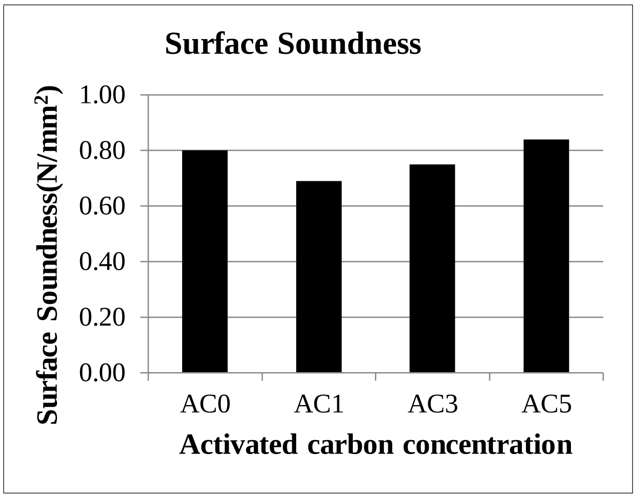

Figure 4. Surface soundness results of MDF panels

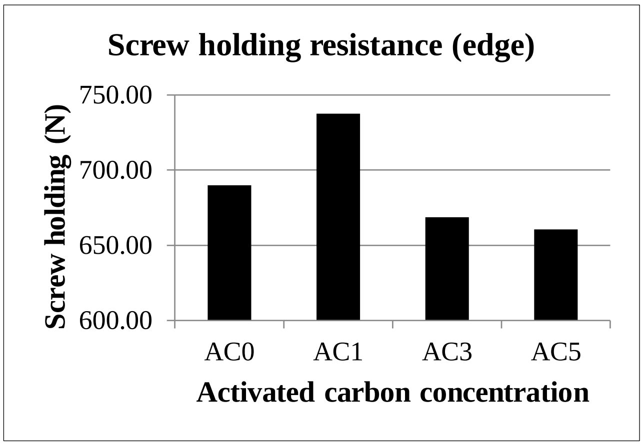

Figure 5. Screw holding resistance results of MDF panels

Mechanical tests were examined; there was a 6.25% increase in internal bonding values for the 1% concentration. With the addition of activated carbon at increasing concentrations for the 3% and 5% ratios, there was a 5.9% decrease and a 12.5% decrease in the internal bonding values, respectively. The screw holding resistance values are analyzed, an increase of 6.4%, a decrease of 3.1% and a decrease of 4.2% were observed, respectively compared to the reference at increasing concentrations. When the surface soundness test results are examined, it is 12.5% decrease, 6.25% decrease, 5% increase compared to reference at increasing rates respectively.

Based on the test results of MDF internal bonding strength, it can be deduced that MDF with less active carbon added mostly exhibits higher strength than control MDF. This can be explained by the fact that the incorporation of activated carbon in MDF fills the space between the fibers in the MDF, thereby intensifying the close contact of the fiber-carbon-fiber system, thereby strengthening the hydrogen bond and van der Waals forces. (Darmawan et al., 2010)

By holding the formaldehyde by the activated carbon, the free formaldehyde in the resin is prevented from escaping from the reaction medium during the curing of the formaldehyde. This strengthens cross-linking. However, the higher activated carbon loading (above 1%), results in less effective retention of formaldehyde due to the agglomeration of the activated carbon particles, the internal bonding strength is reduced. (Resmi et al., 2017)

3.3. Formaldehyde emission tests of MDF panels

Figure 6 shows the formaldehyde emission testing results by the perforator method. The formaldehyde emission tests were done with samples that having 5% moisture content. The value of formaldehyde emission of samples that are named as AC0, AC1, AC3 and AC5 are respectively 22.33, 10.60, 10.67 and 11.78 mg/100g board.

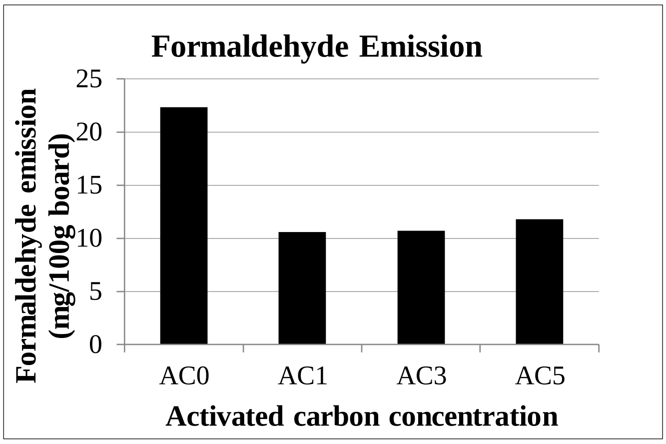

Figure 6. Formaldehyde emission results of MDF panels

According to formaldehyde emission test results, with the addition of activated carbon, a decrease of 52.5%, 52.2% and 47.2% was observed in the emission values, respectively.

That the addition of activated carbon into the resin system reduces the formaldehyde emission values for all concentrations was detected. Such lowering was caused by the capability of the microstructure of the activated charcoal to adsorb formaldehyde in the MDF (Rong et al., 2002; Pari et al., 2006; Beram et al., 2021). Further, the porous structure afforded greater surface area of the adsorbent (activated carbon) and the holding of the adsorbate (formaldehyde) by activated charcoal through the secondary force of hydrogen bonding as well as van der Waals type. This enhanced the intimate take-up of adsorbate on the surface of adsorbent, thereby intensifying the adsorption of formaldehyde by the activated carbon incorporated MDF.

Since the activated carbon used has an iodine number value of over 900 and surface area high that its adsorption capacity to be high based on the information in the literature was expected situation (Medek 2006)

4. Discussion and Conclusions

The main purpose of this study is to produce MDF panels that are sensitive to the environment and human health by reducing formaldehyde emission. For this reason, activated carbon, which is a good adsorbent due to its surface area and porous structure, was used as filler in the urea formaldehyde resin system. The study also aimed to protect the mechanical and physical strength values while reducing the emission values. For this reason, the threshold value for the addition of activated carbon into the resin system has been tried to be determined.

In addition to the emission tests, when all mechanical and physical strength tests were examined, it was decided that the optimum activated carbon concentration was 1%. With the addition of 1% active carbon, the emission value was reduced, while the internal bonding strength, screw holding resistance were increased and a value close to the reference was obtained in surface soundness tests. For thickness swelling and water absorption values, we see that the addition of activated carbon does not cause a significant change compared to the reference.

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