Study of the effect of alfa-x to the urea formaldehyde adhesive on plywood

Kontrplaklarda üre formaldehit tutkalına ilave edilen Alfa-x'in etkisinin İncelenmesi

Yazar(lar) (Author(s)): Murat ÖZALP

ORCID: 0000-0003-1651-3487

Bu makaleye şu şekilde atıfta bulunabilirsiniz(To cite this article): Özlarp M., “Study of the effect of alfa-x to the urea formaldehyde adhesive on plywood”, Politeknik Dergisi, *(*)

Erişim linki (To link to this article): http://dergipark.org.tr/politeknik/archive

DOI: 10.2339/politeknik.769304
Study of the Effect of Alfa-x to the Urea Formaldehyde Adhesive on Plywood

Highlight

❖ It was found that the addition of Alfa-x to the adhesive mixture used on the plywood slightly decreased strength properties of plywood that is bonded using this resin
❖ It was concluded that, since formaldehyde emissions hold a significant importance for human health, Alfa-x, which decrease the levels of this emission

Graphical Abstract
Kontrplaklarda Üre-formaldehit (UF) tutkalına ilave edilen Alfa-x’in formaldehit emisyonu ve mekanik özelliklerini üzerine etkileri incelenmiştir.

Figure. Experimental design

Aim
This study investigated the effect of adding Alfa-x to the urea-formaldehyde (UF) adhesive.

Design & Methodology
The content of free formaldehyde, withdrawal shear strength, and bending strength were studied in the experimental plywood.

Originality
Alfa-x was added to the adhesive (UF) in 0, 10, and 20 wt% ratio of the total amount of the solid content.

Findings
There were higher withdrawal-shear strength and bending strength values of the ekaba plywood than the poplar plywood values. There were higher values of free formaldehyde in the ekaba plywood than the poplar plywood, and the free formaldehyde content value in both samples was reduced by adding Alfa-x to the adhesive.

Conclusion
Formaldehyde emission, which is important for human health, without reducing the mechanical properties of plywood, has been successfully reduced.

Declaration of Ethical Standards
The author(s) of this article declare that the materials and methods used in this study do not require ethical committee permission and/or legal-special permission.
Study of the Effect of Alfa-x to the Urea Formaldehyde Adhesive on Plywood

ABSTRACT

This study investigated the effect of adding Alfa-x to the urea-formaldehyde (UF) adhesive. The content of free formaldehyde and the bonding specifications were examined. The three-layered plywood of poplar (Populus x euramericanus) and ekaba (Tetraberlinia bifoliolata) was prepared by adding alfa-x to adhesive mixtures in specified amounts. The content of free formaldehyde, withdrawal shear strength, and bending strength were studied in the experimental plywood. The strength specifications of plywood bonded with this resin were slightly reduced by adding the Alfa-x to the adhesive mixture. There was also a moderate decrease in the content of free formaldehyde.

Keywords: Content of free formaldehyde, alfa-x, plywood, bending strength, withdrawal shear strength.

1. INTRODUCTION

It is very important to have wood and surfaces in the wood industry. Urea formaldehyde adhesive is generally used because it is cheap and easily available in plywood production [1]. Synthetic resins used in plywood production are substances harmful to human and environmental health. Formaldehyde, which is an important component of synthetic resins, separates from the material at the place of use and mixes with air and creates adverse effects. Formaldehyde has an irritating effect on the eye and nasal mucosa. It can also cause shortness of breath and allergic skin conditions [2].

Formaldehyde spreads from wood-based panels bonded with amino and phenolic resins during production and end use. The amount of formaldehyde released from wood-based panels is affected by exterior factors such as air humidity, air exchange and temperature and production conditions, as well as endogenous factors such as raw material types and resin types [3]. There are several reports about the negative effects of formaldehyde on human health [4,5,6].

Indoor products including core panels of doors, subflooring, and furniture are made using composite wood products including plywood, particleboard, and MDF [7]. One concern for these products is air pollution in residential buildings, which causes health problems. Building materials and construction products cause emit pollutants including acetaldehyde and formaldehyde [8]. The products containing formaldehyde such as plywood, oriented strand board (OSB), resins, insulating material, fabrics, and glues are the main anthropogenic sources affecting humans in closed environments [9]. Large amounts of formaldehyde are manufactured in industry and widely applied. The terpenoid and ozone reaction in the indoor environment produces formaldehyde [10]. For this reason, wood adhesives without formaldehyde should be developed based on industrial safety standards. There should be emphasis on the replacement or reduction of formaldehyde contents in adhesives [11,12,13,14].

A study investigated the volatile acetic acid and formaldehyde emissions effect on the plywood exposed to boric acid and borax. Borax reduces formaldehyde emissions, while boric acid increases them [15]. Any material that creates a panel causes volatile organic compound (VOCs) emissions. Recently, formaldehyde emission from UF adhesive has been examined [16]. To
evaluate the health and environmental effects of the board materials based on wood, formaldehyde emission should be taken into account [17]. An effective and simple approach that suppresses the emission of formaldehyde from plywood can be provided using natural compounds including vanillin, catechin, and urea [18]. The adhesive strength in boards of the laminated veneer lumber (LVL) can be enhanced by adding boric acid to melamine-urea-formaldehyde adhesive [19]. There has been extensive study on the dimensional, biological, and mechanical characteristics of the wood-based materials. According to measurements using the desiccator method, formaldehyde infused with borax decreases formaldehyde emission caused by the plywood panels [20]. Furthermore, there have been studies on the effect of adding borax pentahydrate to urea-formaldehyde (UF) adhesive on the plywood bonding characteristics and its content of free formaldehyde. The plywood withdrawal shear strength and bending strength are not statistically affected by adding borax pentahydrate, but the content of free formaldehyde is reduced [21]. Formaldehyde content of decorative boards based on larch bark (0.6 g/cm³) was analyzed when bonded with five different types of adhesive systems: urea-formaldehyde, polyvinyl acetate, the mixture of 70% urea-formaldehyde + 30% polyvinyl acetate, polyurethane, and tannin-based adhesive. A self-agglomerated board was also analyzed. In the case of boards bonded with tannin-based adhesive, this natural polymer acted as a formaldehyde scavenger [22].

The aim of this study is to investigate the effect of alfa-x in decreasing the free formaldehyde content in urea-formaldehyde adhesive which is used in the production of plywood, and to determine the ideal mixture ratio by researching the effect of the alfa-x that is added into the adhesive mixture in different ratios on the mechanical characteristics of the material.

2. MATERIAL AND METHOD

2.1. Wood material

To produce plywood in experiments, poplar (Populus x euramericana) and ekaba (Tetraberlinia bifoliolata) tree species were used for constructing the veneers. The veneers were made from one lumber for both tree types to ensure the biological and chemical properties of the tree species found in veneers.

2.2. Adhesive

Plywood was produced with urea-formaldehyde (UF) adhesive. Approximately 90% of wooden boards are produced using this plywood, as it requires a short setting term in the hot-press, and it is transparent, easy, and inexpensive. The urea-formaldehyde 2265 adhesive (Polisan Chemical Industry Company, Kastamonu, Turkey) technical properties were as follows: storage for 90 days at 20 °C, viscosity at 20 °C is 100 to 200 (cps), density at 20 °C is 1.22 to 1.23 (g/cm³), rate of solid material (wt%) is 55 ± 1, pH at 20 °C is 7.5 to 8.5, molar ratio (formaldehyde /urea) is 1.35, and free formaldehyde (%) is at most 0.8.

A rolling press was used by applying 180 g/m² of the adhesive double glue line on the sample surface. The test plywood was produced using urea formaldehyde (UF) adhesive with a solid content of 55 wt%. A hardening agent, i.e., 15 wt% of ammonium chloride (NH₄Cl), was added to 10 wt% of the liquid adhesive. An additive of wheat flour was added to the adhesive in 55 wt% of the solid matter.

The Alpha-X antifire is a phosphate-based product with a proprietary formulation. Its unprecedented effectiveness as a flam-retardant is the result of its innovative formulation including also special coupling agents. Usable as an additive in the production process of different materials, it will be homogeneously distributed in the material and imbue it with its unrivaled fire-protection capabilities from the very beginning. It surpasses all requirements demanded for modern flame retardants: absolutely effective, universally applicable and finally safe. The Alpha-X antifire powder includes special titanium derived coupling agents which react with free protons at the inorganic interfere resulting in the formation of organic-titanium mono-molecular layers on the inorganic surface. Typically, tita rate-treated inorganics are hydrophobics, organophilic, and organosurface. Fillers may be pretreated or treated in used in polymers, titanates: increase adhesion; reduce embrittlement; allow higher filler loading; optimize particulate dispersion; increase flow filled and unfilled polymers at lower process temperature; prevent phase separation. The use coupling agents (<10%) are mainly compose of pyrophosphato-O, silicon dioxide and titandioxide.

The Alpha-X antifire is used as a complement to the polymer and is added to the production processes of various products. It is used in thermoplastic, paint, cable, synthetic, membrane, and wood-based boards. It is effective up to 1500 °C against flammability. Alfa-x was added to the adhesive in 0, 10, and 20 wt% ratio of the total amount of the solid content. The technical properties of the Alpha-x are given in Table 1.

| Table 1. Technical Properties of the Alpha-X Chemicals | Property | Specification |
|--------------------------------------------------------|----------|---------------|
| | Appearance | Dry powder |
| | Color | White |
| | Granular size | <5 0 μm |
| | pH | 3.74 |
| | Relative density | 0.847 (g/cm³) |
| | Odor | None |
| | Storage | Cool, dry place; protected from moisture |
| | Web | www.alfa-x.eu (info@eurosis.de) |
2.3. Bending strength

The specifications of BS EN 310 (1993) were followed to analyze the plywood bending strength [23]. The samples measuring 50 mm in width and 150 mm in length were prepared for examination (10 replicates) and the axis of length was in the same direction with the exterior layers fiber direction. Then, the samples were adjusted to the new climate in the air conditioning cabin at relative humidity of 65% and temperature of 20°C. The sample bending strength was calculated with the following method:

\[
\sigma_e = \frac{3fl}{2ba^2}
\]

where \(\sigma_e\) is the bending strength (N/mm\(^2\)), \(f\) is the maximum force at the breakage moment (N), \(l\) is the distance of the support points (mm), \(b\) is the sample piece width (mm), and \(a\) is the sample piece thickness (mm).

2.4. Withdrawal-shear strength

BS EN 314-2 (1993) was followed to determine the withdrawal-shear strength [24]. The samples measured 250 mm in width and 100 mm in length were prepared for examination (10 replicates) such that the axis of length was in the same direction with the exterior layers fiber direction. The samples were adjusted to the new climate in the air conditioning cabin at relative humidity of 65% and temperature of 20°C. The withdrawal-shear strength was calculated with the following equation.

\[
\sigma_w = \frac{F}{A} = \frac{F}{(b.l)}
\]

where \(\sigma_w\) is the withdrawal-shear strength (N/mm\(^2\)), \(F\) is the maximum force at the breakage moment (N), \(b\) is the cross-section sample width (mm), and \(l\) is the cross-section sample length (mm).

The universal test device for mechanical experiments is given in fig.1.

2.5. Free formaldehyde Measurements

The content of free formaldehyde (in 3 replicates) was calculated for measurement of free formaldehyde in the laboratory at the quality control management department of Kastamonu Integrated Company (Kastamonu, Turkey). The TS 4894 EN 120 (1999) was applied for conducting the measurements [25].

The perferator test device in which formaldehyde emission tests are performed is given in fig.2.

2.6. Preparation of experimental samples

The middle layer measuring 2.2 mm and also outer layers ekaba and poplar veneers measuring 1.1 mm were employed to produce a three-layered plywood in size of \(80 \times 80 \times 0.4\) cm. The electric heated hydraulic press with the press area of \(150 \times 300\) cm\(^2\) was applied to press the boards. The press area pressure was set at 10 (kg/cm\(^2\)), and the temperature was 110 °C according to the applicable standards. Pressing lasted for 5 min.

3. RESULTS AND DISCUSSION

Table 2 shows the withdrawal-shear strength and bending strength average values in the ekaba and poplar plywood, which was produced to determine the effect of adding Alfa-x to the adhesive mixture in different ratios.
Table 2. Withdrawal-Shear Strength and the Bending Strength Values (N/mm²)

| Wood Type | Values | Bending Strength Values | Withdrawal-shear Strength Values |
|-----------|--------|-------------------------|---------------------------------|
|           |        | Alfa-x ratio (wt%) | Alfa-x ratio (wt%) |
|           |        | 0      | 10    | 20    | 0     | 10    | 20    |
| Ekaba     | Max.   | 97.68  | 97.38 | 96.94 | 13.15 | 12.48 | 12.12 |
|           | Min.   | 92.64  | 91.52 | 90.83 | 12.01 | 11.87 | 10.99 |
|           | Average| 95.53  | 94.28 | 93.88 | 12.44 | 12.15 | 11.89 |
|           | Stand. Dev. | 1.33 | 1.47 | 1.44 | 0.30 | 0.16 | 0.37 |
| Poplar    | Max.   | 81.12  | 78.47 | 75.15 | 9.75  | 9.78  | 9.61  |
|           | Min.   | 67.00  | 68.41 | 69.98 | 9.34  | 9.13  | 8.98  |
|           | Average| 74.07  | 73.39 | 72.72 | 9.53  | 9.44  | 9.38  |
|           | Stand. Dev. | 3.48 | 2.37 | 1.48 | 0.12 | 0.16 | 0.22 |

There were higher withdrawal-shear strength and bending strength values of the ekaba plywood than the poplar plywood values. However, the withdrawal-shear strength and bending strength values of both samples types were reduced by adding the Alfa-x to the adhesive. Variance analysis was used to examine the interaction between these obtained values, as shown in Tables 3 and 4. For the poplar and ekaba plywood, the variance analysis results showed an insignificant effect of adding the Alfa-x to the adhesive mixture on the bending strength, with a 5% error margin. The effect of adding the Alfa-x to the adhesive mixture on the withdrawal-shear strength was slightly significant, with a 5% error margin.

Table 3. Results of the Bending Strength Variance Analysis

| Source of Variance | Df | Sum of Squares | Mean Squares | F test | P |
|--------------------|----|---------------|--------------|--------|---|
| Tree Type          | 1  | 6723.38       | 6723.38      | 1534.012 | 0.000 |
| Alfa-x ratio (wt%) | 2  | 23.007        | 11.504       | 2.625  | 0.082 |
| Wood*Alfa-x        | 2  | 0.782         | 0.391        |        |    |
| Error              | 54 | 236.67        |              |        |    |
| Corrected total    | 59 | 6983.84       | 4.383        | 0.089  | 0.915 |

Table 4. Results of Withdrawal-Shear Strength Variance Analysis

| Source of Variance | Df | Sum of Squares | Mean Squares | F test | P |
|--------------------|----|---------------|--------------|--------|---|
| Tree Type          | 1  | 110.134       | 110.134      | 1857.433 | 0.000 |
| Alfa-x (wt%)       | 2  | 1.22          | 0.625        | 10.542 | 0.000 |
| Wood*Alfa-x        | 2  | 0.40          | 0.200        | 3.373  | 0.042 |
| Error              | 54 | 3.202         | 0.059        |        |    |
| Corrected total    | 59 | 114.986       |              |        |    |

Table 5 shows the content of free formaldehyde. The content of free formaldehyde in plywood was reduced by adding the alfa-x to the adhesive mixture. There were higher values of free formaldehyde in the ekaba plywood than the poplar plywood, and the free formaldehyde content value in both samples was reduced by adding Alfa-x to the adhesive. Variance analysis was used to examine the interaction between the values, and Table 6 shows the obtained results. There was a relatively significant effect of adding the Alfa-x to the adhesive mixtures in ekaba and poplar plywood on the content of free formaldehyde, with a 5% error margin.
Table 5. The Free Formaldehyde Content Values

| Wood type | Values | Content of formaldehyde (mg/100g) | Alfa-x ratio (wt%) |
|-----------|--------|----------------------------------|-------------------|
|           |        | 0 | 10 | 20 |
| Ekaba     | Max.   | 45.81 | 23.17 | 21.79 |
|           | Min.   | 45.11 | 22.81 | 21.01 |
|           | Average | 45.46 | 22.19 | 21.40 |
|           | Stand. Deviation | 0.35 | 0.18 | 0.40 |
| Poplar    | Max.   | 38.22 | 29.03 | 21.68 |
|           | Min.   | 38.10 | 28.90 | 21.63 |
|           | Average | 38.15 | 28.97 | 21.78 |
|           | Stand. Deviation | 0.06 | 0.06 | 0.13 |

Table 6. Results of Free Formaldehyde Content Variance Analysis

| Source of Variance | Df | Sum of Squares | Mean Squares | F test | P |
|--------------------|----|----------------|--------------|--------|---|
| Tree Type          | 1  | 0.445          | 0.445        | 7.844  | 0.016 |
| Alfa-x ratio (wt%) | 2  | 1357.17        | 678.58       | 11963.37 | 0.000 |
| Wood*Alfa-x        | 2  | 133.42         | 66.71        | 1176.08 | 0.000 |
| Error              | 12 | 0.681          | 0.057        |        |    |
| Corrected total    | 17 | 1491.72        |              |        |    |

4. CONCLUSIONS

Plywood is mandatory in places where robustness and resistance are at stake. In this study, formaldehyde emission, which is important for human health, without reducing the mechanical properties of plywood, has been successfully reduced.

1. There were higher withdrawal-shear strength and bending strength values of the ekaba plywood than the poplar plywood values. It was shown that adding the alfa-x to urea formaldehyde decreased to a small extend the strength characteristics of plywood which is bonded with this resin.

2. Besides, there was a less extreme decrease in content of free formaldehyde. There were higher values of free formaldehyde in the ekaba plywood than the poplar plywood, and the free formaldehyde content value in both samples was reduced by adding Alfa-x to the adhesive.

3. The conclusion was that Alfa-x reducing formaldehyde emissions levels should be used preferably in building material because such emissions are highly important for the human health.

DECLARATION OF ETHICAL STANDARDS

The author(s) of this article declare that the materials and methods used in this study do not require ethical committee permission and/or legal-special permission.

REFERENCES

[1] Wang Y. X., Liu G. Z. and Zhao Y. Q., “China Adhesives”, 17, p.44, (2008).
[2] Coplugil E., “Formaldehyde and formaldehyde resins”, ORENKO-93, 2nd National Forest Products Industry Congress, 6-9 October, Trabzon, K.T.U. Faculty of Forestry, Proceedings, 84-92, (1993).
[3] Boran S., Usta M., Ondaral S., Gümüskaya E., “The efficiency of tannin as a formaldehyde scavenger chemical in medium density fiberboard”, Compos Part B Eng. 43(5): 2487–2491, (2012).
[4] Tang X., Bai Y., Duong A., Smith MT., Li L., Zhang., “Formaldehyde in China: production, consumption, exposure levels, and health effects”, Environ Int 35(8):1210–1224, (2009).
[5] Vosoughi S., Khavanin A., Salehnia M., Mahabadi HS., Soleimanian, A., “Effects of simultaneous exposure to formaldehyde vapor and noise on mouse testicular tissue and sperm parameters”, HealthScope, 1(3): 110–117, (2012).
[6] Norliana S., Abdulamir AS., Abu Bakar F., Salleh AB., “The health risk of formaldehyde to human being”, Am. J. Pharm Toxi, 4(3): 98–106, (2009).
[7] Bodalal A., Zhang J. S., and Plett E. G., “A method for measuring internal diffusion and equilibrium partition...
coefficients of volatile organic compounds for building materials", *Building and Environment* 35(2): 101-110, (2000).

[8] Wanner H.U., “Sources of pollutants in indoor air”, *IARC Scientific Publications*, 109, 19–30, (1993).

[9] Singer B.C., Coleman B.K., Destailhats H., Hodgson A.T., Lundin M.M., Wesceler C.J., Nazaroff W.W., “Indoor secondary pollutants from cleaning product and air freshener use in the presence of ozone”, *Atm. Env.* 40(35): 6696–6710, (2006).

[10] Nicolas M., Ramalho O., Maupetit F., “Reactions between ozone and building products: Impact on primary and secondary emissions”, *Atm. Env.* 41(15): 3129–3138, (2007).

[11] Abdullah Z. A., and Byung D. P., “Influence of acrylamide copolymerization of urea-formaldehyde resin adhesives to their chemical structure and performance”, *Appl. Polym. Sci.* 117(6): 3181-3186, (2010).

[12] Jin Y.Q., Cheng X.S., Zheng Z.B., “Preparation and characterization of phenol-formaldehyde adhesives modified with enzymatic hydrolysis lignin”, *Bioresour Technol* 101(5), 2046–2048, (2010).

[13] Yong S.O., Kyung H.K., “Evaluation of melamine-modified urea-formaldehyde resin for plywood flooring adhesive application”, *Scientia Forestalis* 39(90), 199–203, (2011).

[14] Ozalp M., “Study of the effect of adding the powder of waste PET bottles and borax pentahydrate to the urea formaldehyde adhesive applied on plywood”, *Eur J Wood Prod* 69(3), 369–374, (2011).

[15] Colakoglu G., Colak S., “Volatile acetic acid and formaldehyde emission from plywood treated with boron compound”, *Building and Environment* 39(5), 535–536, (2004).

[16] Baumann M. G. D., Lorenz L. F., Battermann S. A., and Zhang G. Z.; “Aldehyde emissions from particleboard and medium density fiberboard products”, *Forest Products Journal* 50(9): 75-82, (2000).

[17] Sundman M.R., Larsen A., Vestn E., Weibull A., “Formaldehyde emission-comparison of different standard methods”, *Atmospheric Environment* 41(15): 3193–3202, (2007).

[18] Uchiyama S., Matsushima E., Kitao N., Tokunaga H., Ando M., Otsubo Y., “Effect of natural compounds on reducing formaldehyde emission from plywood”, *Atmospheric Environment* 41(38): 8825–8830, (2007).

[19] Bridaux V., Charrier B., Fauroux N., Charrier F., Gonzalez J., “Addition of boron based compound in the LVL glueline: Effect on the mechanical properties and the leaching of boron”, *Holzforschung*, 55(6), (2001).

[20] Colakoglu G., Denirkir C., “Characteristics of plywood panels produced with urea formaldehyde resin containing borax”, *Holz als Roh- und Werkstoff* 64(3): 250-251, (2006).

[21] Sensogut C., Ozalp M., and Yeşil H., “The effect of borax pentahydrate addition to urea formaldehyde on the mechanical characteristics and free formaldehyde content of plywood”, *International Journal of Adhesion and Adhesives* 29(5), 589-592, (2009).

[22] Tudor E.M., Barbu M.C., Petutschnig A.; Réh R. And Krišták L., “Analysis of Larch-Bark Capacity for Formaldehyde Removal in Wood Adhesives”, *Int. J. Environ.Res. Public Health*, 17, 764, (2020).

[23] BS EN 310. “Wood-based panels. Determination of modulus of elasticity in bending and of bending strength”, *British Standards Institution*, London, UK, (1993).

[24] BS EN 314-2, “Plywood - Bonding quality Part 2: Requirements (R)”, *British Standards Institution*, London, UK, (1993).

[25] TS 4894 EN 120, “Wood based sheets boards-Determination of formaldehyde content”, *TSE*, Ankara, Turkey, (1999).