

The Effect of Methods of Introducing Acrylic Emulsions Grafted with Acetoacetyl Groups on the Properties of Particleboards

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ABSTRACT: The aim of this study was to investigate the effect of compounds, either spread on particles or added to the resin, which contain various numbers of acetoacetyl groups in terms of their effect on formaldehyde content and the physical and mechanical properties of particleboards glued with the urea-formaldehyde (UF) resin. The produced particleboards were subjected to the following European Standards standard tests: internal bond, modulus of rupture, modulus of elasticity, swelling in thickness after 24 h of soaking in water, and the content of formaldehyde. Results of the study make it possible to state that both the spreading

of the acrylic emulsions containing acetoacetyl groups on the particles before gluing and the adding of them to the UF resin result in a decrease of formaldehyde content and improvement of physical and mechanical properties of particleboards. Boards with better properties were obtained in the case when emulsions containing acetoacetyl groups were spread directly on the surface of particles. © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 91: 2256–2264, 2004

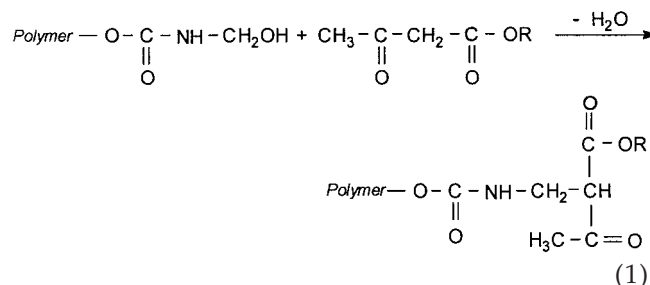
Key words: modification; graft copolymers; acetoacetyl groups; resins; particleboards

INTRODUCTION

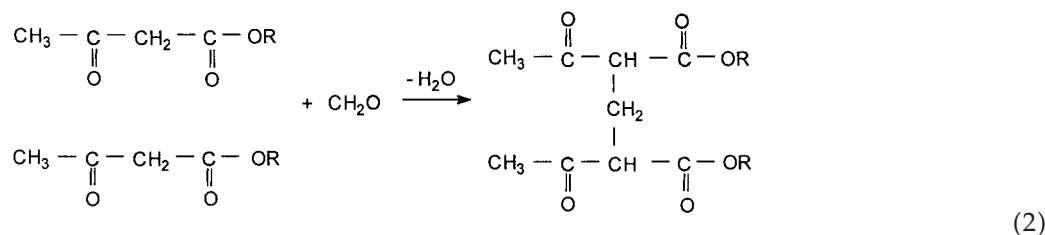
Urea resins are commonly used in the production of particleboards. Yet, boards produced with their use emit significant amounts of formaldehyde. Thus, because of their widespread use in interior furnishings, much research has been aimed at the investigation of the possibilities of limiting the content as well as the emission capacity of this compound. One of the possible methods of restricting CH₂O content may be the use of compounds containing acetoacetyl groups, derivatives of 3-oxobutyric acid CH₃—CO—CH₂COOH.¹

Acetoacetyl groups are amphoteric, and thus, can participate in a variety of chemical transformations which might be used to modify or crosslink poly-

mers.¹ Reagents that are known to react readily with the acetoacetyl groups include amines:



R-alkyl group, aldehydes, especially formaldehyde:



metal ions, and isocyanates.^{2,3}

Therefore, the reactions of amino plastics and formaldehyde with compounds containing acetoacetyl

groups (reactions 1 and 2) led to the formation of crosslinked structures. Moreover, in the former methylene group (presently methine in an acetoacetyl group), there remains an active hydrogen atom which is able to react with free formaldehyde occurring when the particleboards are used. What is more, as a result of the reaction of acetoacetyl groups with hydroxymethylene groups of the resin (reaction 1), car-

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TABLE I
Characteristics of UF Resin (Manufactured by Zakłady
Azotowe Kędzierzyn S. A., Poland)

Kind of determination	Properties of UF resin	
	Actual properties	Properties stated by the manufacturer
Density (g/cm ³)	1.282	—
Dynamic viscosity (mPa s)	610	500–900
No. 4 Ford Cup viscosity (s)	123	60–125
Solids content (%)	67	At least 65
pH	8.09	7.5–8.5
Gel time at 20 °C (h)	Over 6	—
Gel time at 100 °C (s)	86	Max. 110
Miscibility with water	1.2	At least 1.2
Appearance	Milky liquid	—

bon-carbon bonds are formed that hamper the polymer hydrolysis when formaldehyde is emitted due to high temperature or humidity.⁴

It has been reported in literature that compounds containing acetoacetyl groups can be used as formaldehyde-bonding agents, especially in coating systems.^{5–7} Moreover, introducing them to the isocyanates or hydroxyl-functional resins leads to their greater reactivity⁸ and the reinforcement of adhesive-bonded joints.^{9–11} No data have been found on the modification of the urea-formaldehyde (UF) resin by acetoacetyl groups used for the production of particleboards. It is expected that compounds containing acetoacetyl groups, introduced to the UF resin used for the production of particleboards, due to their chemical properties should considerably limit formaldehyde content in particleboards. At the same time, their physical and mechanical properties will improve.

The purpose of the present study is to investigate the effect of compounds containing acetoacetyl groups and the methods of their introduction to the particle-glue mixture on the content of formaldehyde and physical and mechanical properties of particleboards glued with the UF resin.

METHODS

The applied materials

Pure urea-formaldehyde resin, designed for use in the production of particleboards, was applied for experi-

mental purposes. The used UF resin was manufactured by Zakłady Azotowe Kędzierzyn S.A. (Poland). The properties of the UF resin are shown in Table I.

Scotch pine (*Pinus silvestris* L.) particles, obtained under industry conditions, were used to produce particleboards. The particles, before being applied to the production of particleboards, underwent screen classification: dust and the fraction unable to get through square mesh (2 mm on a side) was removed. Some of the particles were ground in a vibratory ball mill; the obtained dust was used in preparing samples used in the determination of activation energy of UF resin crosslinking.

Methyl methacrylate (MMA), acetoacetoxyethyl methacrylate (AAEM), butyl acrylate (BA), styrene (ST), and ethyl-3-ethoxy propionate were purchased from Sigma-Aldrich (Germany). Ammonium chloride (NH₄Cl) and potassium peroxydisulfate (K₂S₂O₈) were purchased from Polskie Odczynniki Chemiczne (Gliwice, Poland). All chemicals were used as received.

Synthesis of acrylic copolymers

The acrylic (AAEM/ST/BA/MMA) copolymers were synthesized by conventional radical polymerization.¹² Ethyl-3-ethoxy propionate solvent was placed in a four-neck round-bottom flask and heated to 100°C. Monomers (AAEM, ST, BA, MMA) and initiator were mixed to complete dissolution. Then, the mixture was placed in an additional funnel and added dropwise to heated solvent over a 4- to 4[1–2]-h period. The initiator was 1.5 mol % K₂S₂O₈. After the addition of the monomer mix was completed, the reaction was continued at 100°C for [1–2] h. To terminate the reaction, an additional [1–2] mol % initiator as a 10% solution in the ethyl-3-ethoxy propionate solvent was added dropwise over a 10- to 15-min period. Then, the mixture was cooled and transferred to a storage container.

The obtained emulsions contained 0, 2412, 1212, and 926 gram-equiv of acetoacetyl groups, respectively, where a gram-equiv of acac groups means the number of grams of the polymer containing 1.0 mol of acetoacetyl groups. The properties of the obtained emulsions are shown in Table II.

TABLE II
Characteristics of Acrylic Emulsions Grafted with Acetoacetyl Groups

AAEM/ST/BA/MMA (mol)	0/40/40/20	12.5/35/35/17.5	25/40/15/20	30/30/20/20
Gram-equivalent of acac groups (g/mol)	0	2412	1212	962
pH	7.20	7.14	7.06	7.00
No. 4 Ford Cup viscosity (s)	76	70	65	52
Dynamic viscosity (mPa s)	240	195	160	140

The effect of acrylic emulsions containing acetoacetyl groups on UF resin at 20°C

The use of acrylic emulsions that activate the curing process leads to significant changes in the viscosity of the prepared mixed glue. Thus, it was possible to determine the effect of acrylic emulsions added to the UF resin on its viscosity at 20°C as a function of time. To achieve this purpose, immediately before the determination, solutions of mixed glue were prepared, containing, respectively, 0, 2.5, 5, 7.5, and 10 parts by weight of appropriate emulsions per 100 parts by weight of dry resin solids. The dynamic viscosity was determined every 2 h for 12 h, and then every 12 h. The measurements were completed after 60 h. The dynamic viscosity of the solution was determined with a Höppler rheoviscometer (VEB MLW Prüfgeräte Werk Medingen, Dresden) (The name Höppler rheoviscometer is a brand name used by the manufacturer. However, the referred viscometer is in fact a falling-weight viscometer.)

Gel time of UF resin modified with acrylic emulsions and determination of the activation energy of crosslinking

The ability to activate the thermosetting resins with various additives is commonly estimated by measurements of the gel time of the mixed glue at different temperatures. Therefore, the gel times were examined at the temperatures of 80, 90, 100, and 110°C. The solutions of mixed glue were prepared by adding appropriate emulsions to the resin immediately before the determination. They consisted of, respectively, 0, 2.5, 5, 7.5, and 10 parts by weight of the appropriate emulsions per 100 parts by weight of dry resin solids. The amount of curing agent was two parts by weight per 100 parts by weight of dry resin solids. To eliminate the effect of the pH changes on the gel time of the mixed glue, the pH values of the modified UF resins were measured at 20°C. The pH of the solutions was determined with a pH-meter CP-501 (Elmetron, Poland).

Moreover, to determine the possible effect of acrylic emulsions on the reaction of UF resin toward wood, the following mixtures were prepared and tested: resin and curing agent; resin, curing agent, and appropriate emulsions; resin, curing agent, and pine dust; resin, curing agent, appropriate emulsions, and pine dust. The amount of acrylic emulsions was 10 parts by weight per 100 parts by weight of dry resin solids. The amount of curing agent was two parts by weight per 100 parts by weight of dry resin solids. The amount of pine dust was 10 parts by weight per 100 parts by weight of dry resin solids.

The gel times of mixed glue were used to assess the relative activation energy of the crosslinking process of the tested variants.¹³ A preliminary assumption

was made that the curve illustrating the curing process in time, for the particular temperatures of 80, 90, 100, and 110°C, was a linear function until the curing point was reached. Because the reaction rate increases along with the growth of the temperature, and the curve illustrating the curing rate, in dependence of the inverse of the absolute temperature, shows a very good linear correlation, the activation energy (E_a) can be calculated based on the Arrhenius equation:

$$k = A \exp(-E_a/RT)$$

where k is the curing reaction rate constant, A is the preexponential factor, R is the gas constant, and T is the absolute temperature.

The production of laboratory particleboards and the examination of their properties

To examine the effect of the compounds containing acetoacetyl groups on the properties of particleboards glued with UF resin, single-layer boards with the density of 700 kg/m³ and dimensions of 500 × 600 × 12 mm were produced under laboratory conditions. The following pressing parameters were employed: pressure, 2.5 MPa; temperature, 180°C; time, 5 min. Their resin content was 10% by oven-dry wood weight. The appropriate emulsions were either added to the resin or spread directly on the particles before gluing in the amounts of 2.5, 5, 7.5, and 10% in relation to dry resin solids. For each variant, three boards were made. The acrylic emulsions, urea resin, and their mixture were spread over the particles in a slow-speed laboratory blender.

The used curing agent was 20% solution of ammonium chloride in the amount of 2% in relation to dry resin solids.

The properties of the particleboards were examined according to the relevant European Standards (EN): modulus of rupture (MOR) and modulus of elasticity (MOE)¹⁴; internal bond (IB)¹⁵; swelling in thickness after 24 h¹⁶; and formaldehyde content.¹⁷ In the case of IB, MOR, and MOE evaluation, 12 samples per each board were tested to evaluate the mean value (\bar{x}) and standard deviation (σ).

RESULTS

The effect of acrylic emulsions on resin stability at 20°C and the curing kinetics

The effect of acrylic emulsions containing acetoacetyl groups upon the UF resin stability at 20°C is shown in Table III. The included data indicate that, at first, as the number of acetoacetyl groups in the emulsions grows, the dynamic viscosity of the mixed glue decreases. In the initial 10 h, the viscosity of the solutions

TABLE III
The Effect of Time on the Viscosity of UF Resin Modified with Acrylic Emulsions Containing Acetoacetyl Groups

Amount of modifier (%)	Kind of modifier	Dynamic viscosity (mPa s)									
		Measurements made in time (h)									
		0	2	4	6	8	10	12	24	36	48
0	Control sample	610	610	610	610	615	620	635	650	676	720
2.5	Gram-equivalent of acac ^a groups in emulsion 0 g/mol	465	465	465	470	510	565	605	780	930	1135
5		450	450	450	460	500	555	595	765	910	1175
7.5		420	420	420	430	470	525	565	750	890	1220
10		415	415	415	425	465	520	560	730	875	1240
2.5	Gram-equivalent of acac groups in emulsion 2412 g/mol	390	390	390	420	510	620	975	1090	1930	2950
5		370	370	370	400	495	605	965	1115	2150	3180
7.5		350	350	350	380	470	560	950	1185	2280	3295
10		335	335	335	365	455	545	920	1210	2390	3315
2.5	Gram-equivalent of acac groups in emulsion 1212 g/mol	325	325	330	380	470	560	905	1125	2160	3385
5		310	310	320	375	455	540	885	1200	2300	3445
7.5		300	300	315	365	440	525	875	1230	2385	3490
10		280	280	300	350	430	505	850	1350	2490	3595
2.5	Gram-equivalent of acac groups in emulsion 962 g/mol	240	245	260	310	390	460	860	1295	2405	5350
5		220	225	245	285	365	450	855	1320	2450	5460
7.5		190	195	220	270	350	425	815	1395	2560	5570
10		170	175	205	260	340	420	820	1465	2690	5690

^a Acac, acetoacetyl.

changes slightly. A considerable diversification of the viscosity of the modified and nonmodified UF resin is observed after 24 h. After this time, the dynamic viscosity of UF resin modified with acrylic emulsions increases rapidly. The increase is greatest in the case of emulsions containing the largest number of acetoacetyl groups. It seems that after 24 h the crosslinking process of the UF resin containing acetoacetyl groups intensifies and that results in the increase in the resin viscosity. The decrease in the viscosity observed initially has also been noticed by other researchers.^{18,19} In their studies, the researchers prove that the presence of acetoacetyl groups in amine coating resins significantly decreases its viscosity and, simultaneously, shortens its gel time at ambient temperatures. The initial decrease in the viscosity of the modified UF resins can be explained by the introduction of AAEM,

as the carrier of acetoacetyl groups, into the resin structure: AAEM makes the individual chains of urea polymers more distant from one another. This steric hindrance, as well as the fact that AAEM does not include hydroxyl groups, minimizes the possibility of forming hydrogen bonds between the individual chains of the polymer. As a result, the mobility of polymer chains increases and, therefore, the viscosity of the solution decreases.²⁰

Table IV illustrates the effect of the kind and amount of emulsions introduced into UF resin on the gel time of the resin. The data included in the table prove that the applied modifiers do not affect the pH and slightly decrease the gel time at 100°C. It has been found that the fraction of acetoacetyl groups in the acrylic emulsion is more significant than the amount of the emulsion introduced into the solution. Never-

TABLE IV
Gel Times of UF Resin Modified with Acrylic Emulsions with Various Numbers of Acetoacetyl Groups at 100°C

Amount of modifier in relation to dry resin solids (%)	Investigated formula									
	UF resin		UF + emulsion 0 acac groups		UF + emulsion 2412 acac groups		UF + emulsion 1212 acac groups		UF + emulsion 962 acac groups	
	Gel time (s)	pH ^a	Gel time (s)	pH	Gel time (s)	pH	Gel time (s)	pH	Gel time (s)	pH
0	83	6.68	—	—	—	—	—	—	—	—
2.5	—	—	87	6.47	75	6.46	69	6.48	64	6.52
5.0	—	—	85	6.45	69	6.47	65	6.47	59	6.48
7.5	—	—	83	6.47	66	6.45	62	6.51	49	6.48
10	—	—	81	6.44	65	6.52	60	6.53	47	6.46

^a pH was measured at 20°C.

TABLE V
The Effect of the Wood Dust Addition and the Kind of Acrylic Emulsion Upon Gel Times of UF Resin at Temperatures Ranging from 80 to 110°C

Curing temperature (°C)	Investigated formula									
	UF resin	UF resin + dust ^a	UF resin + emulsion ^b 0 acac groups	UF resin + emulsion 0 acac groups + dust	UF resin + emulsion 2412 acac groups	UF resin + emulsion 2412 acac groups + dust	UF resin + emulsion 1212 acac groups	UF resin + emulsion 1212 acac groups + dust	UF resin + emulsion 962 acac groups	UF resin + emulsion 962 acac groups + dust
80	410 ^a	208	395	186	368	182	318	178	282	172
90	202	101	186	90	164	88	141	88	121	86
100	87	48	83	42	80	41	76	40	70	41
110	54	42	54	38	50	36	48	36	44	37

^a The amount of the added pine dust was 10% of dry mass in relation to dry resin solids.

^b The amount of the added emulsion was 10% in relation to dry resin solids.

^c Numbers are Gel time in seconds.

theless, the greatest shortening of the gel time, by almost 50%, was obtained for the maximum (10%) addition of the modifier, containing the largest number of gram-equivalents of acetoacetyl groups. This fact confirms the chemical reactivity of acetoacetyl groups toward the UF resin.

Table V illustrates how the amount of acetoacetyl groups introduced along with the acrylic emulsion into the resin and the presence of wood dust affect the gel time of the UF resin at the temperatures ranging from 80 to 110°C. Data included in the table prove that introducing acetoacetyl groups into the UF resin considerably shortens the gel time of the resin, especially in lower temperatures. The maximum shortening of the gel time of the UF resin with the addition of 10% emulsion containing the largest number of acetoacetyl groups at 80°C reaches 30%, and the largest number of acetoacetyl groups at 110°C reaches only 19%. Introducing pine dust into the resin also significantly shortens the gel time of the resin; also in this case the effects are less apparent as the temperature grows. The short-

ening of the gel time is most apparent when pine dust and acrylic emulsions are simultaneously introduced into the resin; best results are obtained for emulsions with the largest number of acetoacetyl groups. Nonetheless, the addition of pine dust decreases the gel time of UF resin in a more effective way (Table V).

Table VI shows the effect of emulsions containing acetoacetyl groups and wood dust upon the activation energy of the polycondensation process of UF resin, calculated on the basis of the gel time measurements. The data included in the table show that introducing wood dust along with the used modifiers into the resin makes it possible to decrease the activation energy of the process by as much as 30%.

Results of studies on particleboard properties

The results of studies on the effect of the methods of introducing acetoacetyl groups in acrylic emulsions used as the modifiers of UF resin in the production of particleboards are presented in Tables VII and VIII

TABLE VI
Kinetic Parameters of the Curing Process of UF Resin Modified with Acrylic Emulsions with Various Contents of Acetoacetyl Groups and Wood Dust

Formula	Curing temperature (1000/T)					Activation energy (kJ/mol)	ln A	
	°C	80	90	100	110			
	K	383	393	403	413			
	K ⁻¹	2.61	2.54	2.48	2.42			
		ln k						
UF resin		5.43	5.84	6.74	7.30	80.6	33.3	
UF resin + emulsion 0 acac groups		5.43	5.81	6.66	7.21	76.7	32.0	
UF resin + emulsion 2412 acac groups		5.45	6.00	6.76	7.40	76.5	32.1	
UF resin + emulsion 1212 acac groups		5.46	5.93	6.70	7.26	72.9	30.8	
UF resin + emulsion 962 acac groups		4.99	5.93	6.54	7.26	72.8	30.7	
UF resin + dust		5.66	5.93	6.70	7.26	73.3	30.9	
UF resin + emulsion. 0 acac group + dust		5.37	5.82	6.54	7.01	65.1	28.0	
UF resin + emulsion 2412 acac group + dust		5.40	5.96	6.61	7.09	61.8	27.0	
UF resin + emulsion 1212 acac group + dust		5.37	5.93	6.54	6.59	55.6	24.9	
UF resin + emulsion 962 acac group + dust		5.40	5.83	6.51	6.83	55.1	24.7	

TABLE VII
The Effect of Acrylic Emulsions with Varying Numbers of Acetoacetyl Groups Spread on the Particles on Physical and Mechanical Properties of Particleboards

Amount of emulsion (%)	IB (MPa)		MOR (MPa)		MOE (MPa)		Swelling in thickness after 24 h (%)	
	\bar{x}	σ	\bar{x}	σ	\bar{x}	σ	\bar{x}	σ
Control board	0.55	0.03	14.9	0.3	2230	130	45.7	1.8
Gram-equivalent of acac groups—0 g/mol								
2.5	0.56	0.03	14.8	0.6	2280	100	41.0	1.5
5.0	0.58	0.03	14.9	1.0	2310	100	40.2	3.5
7.5	0.59	0.06	15.4	0.9	2510	190	38.6	3.9
10	0.62	0.03	15.7	1.1	2600	160	34.0	3.4
Gram-equivalent of acac groups—2412 g/mol								
2.5	0.62	0.03	15.7	0.4	2580	170	39.8	2.3
5.0	0.64	0.03	16.0	1.0	2670	110	36.6	2.0
7.5	0.72	0.03	16.9	1.0	2780	130	35.1	1.5
10	0.78	0.03	17.3	0.9	2840	150	32.8	1.9
Gram-equivalent of acac groups—1212 g/mol								
2.5	0.75	0.04	16.9	0.4	2730	80	33.8	2.4
5.0	0.79	0.03	17.5	0.6	2890	150	31.2	2.2
7.5	0.82	0.05	18.1	1.2	3010	150	29.5	2.7
10	0.83	0.03	18.8	0.5	3070	70	27.1	1.5
Gram-equivalent of acac groups—962 g/mol								
2.5	0.83	0.03	17.9	0.3	2980	130	29.8	1.8
5.0	0.85	0.04	18.6	0.8	3140	190	27.2	1.5
7.5	0.91	0.04	19.1	1.1	3330	180	24.3	1.2
10	0.95	0.05	19.4	0.9	3580	200	26.1	1.1

and Figures 1 and 2. Data included in Table VII indicate that spreading the acrylic emulsions directly on the particles, both those containing and not containing acetoacetyl groups, results in the improvement of mechanical and physical properties of particleboards. These values considerably increase, with increasing amounts of the emulsions and their acetoacetyl group content, spread on the particles. It is especially noticeable in the case of the increase of the strength properties with an increase in the amount of acetoacetyl groups being introduced. Thus if, for the 10% addition of acrylic emulsions not containing acetoacetyl groups, the internal bond increases by 13%, then, in the case of adding the emulsion containing the maximum number of acetoacetyl groups, the increase reached 73%. At the same time, the modulus of rupture increased by 5 and 30%, respectively. An increase in the modulus of elasticity by 60% was found in relation to the control board. Moreover, the applied emulsions significantly affect the hydrophobic properties of the particleboards being expressed by the decrease in the swelling in thickness after 24 h. The greatest improvement of the hydrophobic properties,

by almost 43%, was obtained for the emulsion containing the largest number of acetoacetyl groups spread on the particles in the amount of 10%.

As can be seen from Figure 1, spreading the emulsions containing acetoacetyl groups on the particles before gluing limits the formaldehyde content in particleboards: the greater the amount of the acetoacetyl groups the better are results achieved. A decrease in the release of formaldehyde content can be explained by the fact that acetoacetyl groups are capable of bonding free formaldehyde present in the UF resin (according to reaction 2).

A different tendency is observed in the case of spreading, on the particles, the emulsions containing acetoacetyl groups together with the UF resin. Data presented in Table VIII indicate that the improvement of mechanical properties of particleboards is proportional to the amounts of acrylic emulsions, either groups that do not contain acetoacetyl or that contain their least amount, introduced to the resin. Nevertheless, in the case of spreading, together with the UF resin, the acrylic emulsions containing more amounts of acetoacetyl groups (gram-equivalent of acetoacetyl

TABLE VIII
The Effect of Acrylic Emulsions with Varying Numbers of Acetoacetyl Groups Added to the Resin on Physical and Mechanical Properties of Particleboards

Amount of emulsion (%)	IB (MPa)		MOR (MPa)		MOE (MPa)		Swelling in thickness after 24 h (%)	
	\bar{x}	σ	\bar{x}	σ	\bar{x}	σ	\bar{x}	σ
Control board	0.55	0.03	14.9	0.3	2230	130	45.7	1.8
Gram-equivalent of acac groups—0 g/mol								
2.5	0.52	0.04	15.1	0.8	2410	120	39.4	2.1
5.0	0.54	0.05	15.3	1.3	2490	150	38.3	1.8
7.5	0.54	0.05	16.0	1.4	2600	170	37.3	2.0
10	0.56	0.04	16.3	1.3	2640	170	33.8	2.5
Gram-equivalent of acac groups—2412 g/mol								
2.5	0.57	0.03	15.3	1.2	2500	180	43.0	1.1
5.0	0.60	0.06	15.6	1.3	2600	180	42.1	1.0
7.5	0.62	0.06	16.2	1.1	2710	90	39.2	2.1
10	0.64	0.06	16.6	1.4	2790	150	36.3	1.6
Gram-equivalent of acac groups—1212 g/mol								
2.5	0.60	0.06	15.6	1.3	2550	140	49.8	1.9
5.0	0.67	0.05	16.5	1.5	2870	170	44.3	0.9
7.5	0.72	0.06	17.9	0.7	2990	170	39.8	0.9
10	0.68	0.08	17.5	1.7	2880	150	36.5	0.8
Gram-equivalent of acac groups—962 g/mol								
2.5	0.68	0.04	15.9	0.8	2610	180	54.6	3.3
5.0	0.70	0.04	17.0	1.1	2890	220	51.9	2.9
7.5	0.78	0.05	18.8	1.4	3050	190	45.4	2.8
10	0.75	0.04	18.1	1.3	2940	200	40.9	2.4

groups of 1212 and 962 g/mol) optimum strength properties are observed for their addition in the amount of 7.5%.

The swelling in thickness of the produced particleboards is lower than for the control board; however, these values increase, as the amount of added

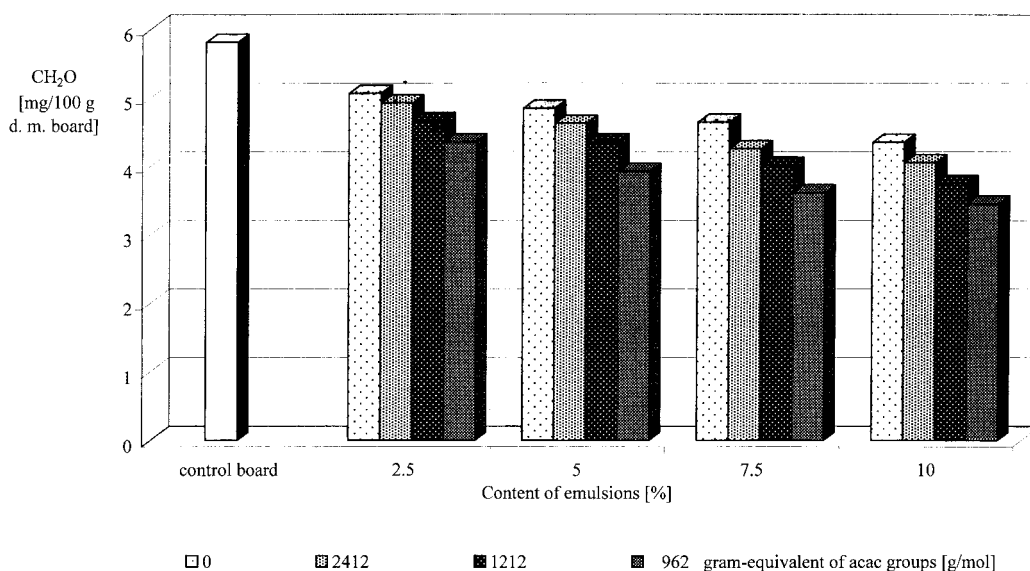


Figure 1 The effect of acrylic emulsions with varying numbers of acetoacetyl groups spread on the particles on the formaldehyde content in particleboards.

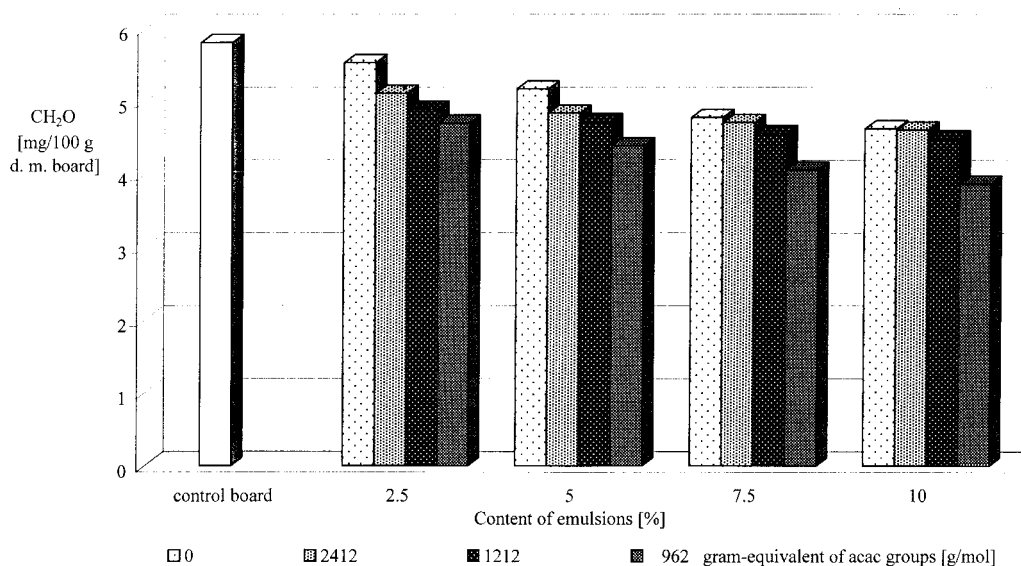


Figure 2 The effect of acrylic emulsions with varying numbers of acetoacetyl groups added to the resin on the formaldehyde content in particleboards.

emulsions and acetoacetyl groups content grow. It may be due to the reaction with the UF resin of acetoacetyl groups contained in the acrylic emulsions. The polycondensation process is then interrupted and, consequently, fewer resin-wood bonds are formed, thus, facilitating water penetration into the particleboards.

Moreover, this method of introducing acetoacetyl groups to the particle-glue mixture results in a decrease of formaldehyde content in the produced particleboards (Fig. 2). Nevertheless, the decrease in formaldehyde content is less effective than in the case of spreading acrylic emulsions on the particles before gluing (decrease in formaldehyde content by 33 and 41%, respectively, for the maximum amounts of acetoacetyl groups in the emulsions). It is expected that acetoacetyl groups introduced in the former case may react in close contact with the UF resin already at the first stage of polycondensation. Hence, their influence on nonreacted hydroxymethylene groups in the polycondensed UF resin is less effective.

CONCLUSION

Introducing acrylic emulsions containing acetoacetyl groups into the liquid UF resin results in the decrease of the resin stability at ambient temperature and an increase in the reactivity under pressing conditions. The shortening of the gel time, and the decrease of the activation energy of the polycondensation process, shows the latter.

Both spreading the acrylic emulsions containing acetoacetyl groups on the particles before gluing and

adding them to the UF resin result in a reduced formaldehyde content in the produced particleboards. A greater reduction of formaldehyde content was obtained in the case when the emulsion was spread directly on the particles.

Either modification of UF resin with the use of acrylic emulsions containing acetoacetyl groups or spreading the emulsions on the particles result in the improvement of physical and mechanical properties of particleboards, especially of internal bond, modulus of rupture, and swelling. These values considerably increase along with the increasing amount of the added emulsions and their acetoacetyl group content. The boards exhibiting better properties were obtained when the emulsions containing acetoacetyl groups were spread directly on the particles.

The results of the studies led to the conclusion that boards with the best physical and mechanical properties and a considerably limited formaldehyde content may be obtained in the case when the emulsion containing 962 g-equiv of acetoacetyl groups in the amount of 10% in relation to the dry resin solids was spread directly on the particles.

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