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WATER-BASED AIR-DRYING ALKYD POLYESTER RESINS MODIFIED WITH GLYCEROL ALLYL ETHER

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Key Words: Polyester alkyd resin; Water thinnable resin; Glycerol allyl ether; Air-drying lacquers

ABSTRACT

A water thinnable polyester alkyd resin containing glycerol allyl ether (GAE) was synthesized. The influence of the content of GAE for various degrees of molecule branching, expressed by the functionality of hydroxylic components, on the properties of the resin and coatings obtained from it was studied. It was found that the lacquers obtained from the alkyd resin containing 10–16 wt% GAE exhibit short drying time, good hardness of coatings, and good water resistance.

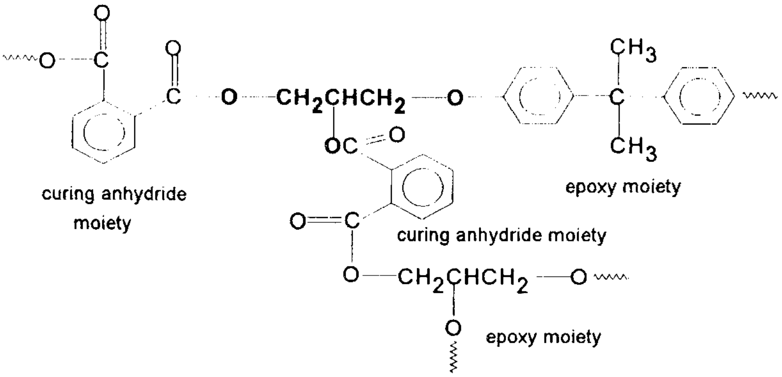
INTRODUCTION

Coating materials based on air drying alkyd polyester resins still remain a high volume group of products. Such coating materials presently produced contain a large amount of organic solvents. This causes many threats, mainly by air pollution. Due to this, there is a trend to obtain new coatings, soluble or dispersible in water, which are not only harmless for the environment but also require a cheap and easily accessible solvent. Additionally, auxiliary solvents used in water-based systems, such as butoxyethanol, due to their higher oxygen content more easily undergo degradation and do not contaminate the environment to such a degree as xylene or petroleum spirits [1, 2].

Water exhibits a number of specific properties requiring modification of binders used for water-based coatings. The high evaporation heat and relatively high freezing point are among these properties. This causes an elongation of the drying time [3] (especially under high air humidity conditions) and requires heated space for storing water-based coatings during winter. On the other hand, due to the large number of ester groups in the macromolecules, alkyd polyester resins exhibit a tendency toward hydrolysis, especially in aqueous alkaline solutions. As a result of the decomposition of macromolecules, e.g., by the abstraction of phthalic acid half esters, the molecular weight decreases as does the number of unsaturated bonds in the molecule, and thus the drying ability [4–7].

Many ways of counteracting this phenomenon have been evaluated, e.g., by using sulfonate groups [8, 9], eliminating ionogenic groups, or incorporating groups resistant to hydrolysis [4, 10]. In order to transfer the alkyd resins into a water-soluble form without the participation of ionogenic groups, poly(oxyethylene) units have been introduced. They can be incorporated into the molecule or added in the form of specially produced resins—emulsifiers. A large number of poly(oxyethylene) units may, however, worsen the coatings properties, among others the resistance toward water. The grafting of acrylic monomers, including acrylic and methacrylic acids, has also been used. The ionogenic groups introduced by this way are linked with the resin molecules via nonhydrolyzable bonds [11–16]. The tendency toward gelation during production as a result of high functionality is a serious disadvantage of such systems [4].

Taking advantage of experience with anhydride-cured epoxy resins [17] whose chemical structure involves a glycerol derivative with two ester and one ether bonds (Scheme 1) assures high resistance toward hydrolysis, we decided to use the glycerol monoallyl ether (GAE), $HOCH_2CH(OH)CH_2{-}O{-}CH_2CH{=}CH_2$, for the modification of the alkyd polyester resin. This monomer contains two hydroxyl groups reactive with carboxylic acids besides an allyl ether group capable of drying in air. Thus, the unsaturated allyl group capable of polymerization, contrary to the fatty acid residues, would be linked with the resin molecule via an ether bond



SCHEME 1.

resistant toward hydrolysis. The substitution of a part of the soybean oil fatty acid moieties (18 carbon atoms) by a small allyl ether molecule (3 carbon atoms) would also enable the introduction of a larger number of reactive unsaturated bonds at almost the same molecular weight, and this might shorten the drying time and increase the hardness of the coating.

Reactive allyl groups may also be introduced into the water-soluble resin by using pentaerythritol allyl ether [15] or allyl glycidyl ether [18]. Surfactants containing allyl ethers as reactive functional groups have been used as copolymerizable emulsifiers for alkyds [19].

The purpose of this work was to study the influence of the content of the incorporated GAE for various degrees of molecular branching, expressed by the functionality of hydroxyl components, on the properties of the alkyd polyester resin and the coatings obtained from it.

EXPERIMENTAL

Resin Synthesis Using Glycerol Allyl Ether

Esters of pentaerithritol and glycerol with soybean oil fatty acids containing free hydroxyl groups (**1**) were used as the resin hydroxyl components besides GAE. Phthalic anhydride and 3,3′,4,4′-benzophenonetetracarboxylic dianhydride (**2**) were used as the acid components. The synthesis was carried out in two steps. Phthalic anhydride, **1**, and GAE in appropriate amounts (maintaining constant average functionalities of the hydroxylic components of 2.28, 2.32, or 2.36, respectively) were placed in a reaction vessel equipped with a stirrer, reflux condenser, thermometer, and Barrett-type receiver. The content of the vessel was subjected to condensation at 180-200°C under nitrogen flow. The condensing water was removed as an azeotrope with toluene. Depending on the average functionality of the hydroxyl components (f_{OH}), the first step was carried out until the acid number of the reaction mixture reached 20-24 mg KOH/g (for f_{OH} = 2.28 and 2.32) or 25-30 mg KOH/g (for f_{OH} = 2.36). After reaching the required acid number, toluene was removed at 220-240°C and the viscosity of the precondensate was measured. The Barrett-type receiver was then exchanged for a reflux condenser, and **2** was added at 120-130°C in such an amount that the final acid number of the resin was roughly 40 mg KOH/g. In order to avoid condensation of the potentially tetrafunctional dianhydride **1**, it was introduced with ca. 4 wt% water. The content of the reaction vessel was heated at 190 ± 2°C until the final acid number of ca. 40 mg KOH/g was reached. The prepared resin was diluted to 85 wt% with a mixture of *n*-butanol and diethylene glycol monobutyl ether (1:2 w/w), and then the dynamic viscosity was measured.

Preparation of the Lacquer

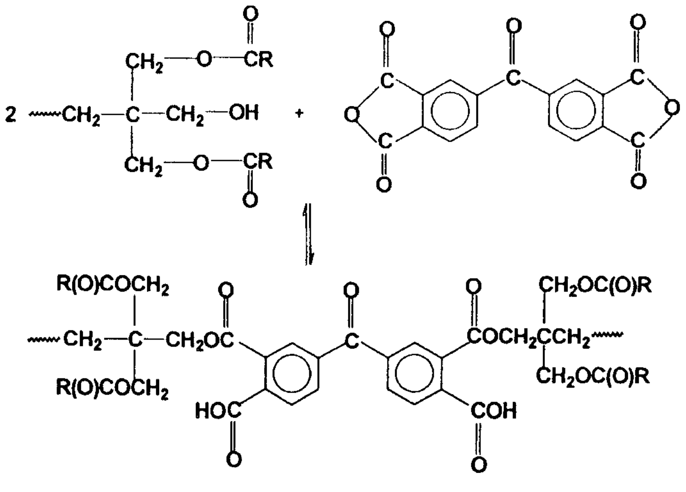
The resin solutions obtained were neutralized with triethylamine (TEA) or a mixture of TEA with an aqueous ammonia solution to pH 8.2. A dryer composed of cobalt naphthenate (0.15% w/w metal content) and zirconium (0.15%) and calcium (0.15%) siccatives was added. The compositions obtained were diluted with demineralized water to 50 and 60% of the dry mass. The prepared composition was applied by means of a bar-coater (thickness of wet layer = 90 μm) onto glass plates, the drying time and coating hardness were measured, and the resistance toward water was studied.

Methods of Investigation

The dynamic viscosities of the resins studied were measured by means of a Mettler-Toledo RM80 rotational viscometer. Molecular weight determinations were carried out by means of a vapor pressure osmometer in chloroform solutions. First degree drying time measurements consisted of recording the time after which quartz sand of the respective sieve fraction (ca. 0.1–0.355 mm grain size) dropped from a height of 10 cm on a lacquer coating applied on a glass plate was easily shaken off or removed by a soft brush without sticking to the surface (Polish standard PN-79/C-81519). Third degree drying time measurement was carried out by placing a square piece of class III copying paper (area: 3 × 3 cm^2) on the drying coating, and placing a 2 cm (in diameter) rubber disk on top of it. Then a 200-g weight was placed on the disk for 1 minute. The third degree drying time was reached when the paper came off easily after removal of the load and did not leave noticeable traces. Measurements of relative hardness (expressed in seconds) of the coatings were carried out by means of a König pendulum (Polish standard PN-79/C-81526). The resistance to water was measured according to the Polish standard PN-79/C-81521.

RESULTS AND DISCUSSION

The use of **2** permitted us to obtain resins of high molecular weight as well as of good solubility in water. Carrying out the reaction with **2** under reflux in the presence of a small amount of water assured an addition mode of reaction—mainly the anhydride groups react with the resin's hydroxyl groups (Scheme 2). An excess



SCHEME 2.

of water vapor above the reaction mixture surface hinders condensation and makes the potentially tetrafunctional dianhydride a bifunctional reagent. Such a procedure yields an oligoester with carboxylic side groups, assuring very good water solubility of the resin after neutralization by an amine.

Resin synthesis with a different amount of GAE was carried out with a constant molar ratio of OH groups to COOH groups in the precondensate (equal to 1.3) and a hydroxylic component functionality equal to 2.28, 2.32, or 2.36 OH groups per molecule. The GAE functionality is equal to 2. Therefore, in order to maintain the required extent of oligoester branching (average functionality of the hydroxylic components), the pentaerithritol and soybean oil reaction product (**1**) charge was enriched with pentaerithritol when the ether share in the resin increased (Tables 1-3).

The increase in GAE content was accompanied by a remarkable increase in the viscosity of the precondensates (Fig. 1), leading in extreme cases (>16 wt% GAE and $f_{OH} = 2.36$) to their gelation prior to reaching the required acid number. In these cases the condensation was carried out at lower conversions (higher precondensate acid numbers). The acid number of precondensates with an average functionality of 2.28 and 2.32 ranged between 20 and 24 mg KOH/g. This means that reaching the same acid number for resins with a larger GAE content, and hence a larger phthalic anhydride content (Tables 1-3), occurred at a higher conversion of carboxylic groups. The molecular weights of the resins also increased. However, the molecular weights of resins with an average functionality of 2.32 were higher than those of resins with $f_{OH} = 2.28$. This was confirmed by viscosity and molecular weight determinations. For example, the molecular weight of resin 4-3 containing 9.4 wt% GAE ($f_{OH} = 2.28$) was 2150 g/mol (for $f_{OH} = 2.32$–3370 g/mol, Table 2), while for resin 8-3 containing 26.4 wt% GAE a molecular weight of 6000 g/mol was found for the same precondensate acid number of 22 mg KOH/g (Table 3).

An increase in the glycerol allyl ether content, and thus an increase in the number of unsaturated bonds of an oligoester molecule, has only a slight effect on shortening the drying time but causes a considerable increase in the hardness of the coating. This relationship can be noticed when analyzing the dependence of hardness on the GAE content (Fig. 2). Characteristic maxima occur here for 16 wt% GAE ($f_{OH} = 2.28$) and 9 wt% GAE ($f_{OH} = 2.36$). Resins with the highest functionality and high GAE content exhibit the worst hardness and resistance to water properties of the lacquers (Tables 4-6).

The course of coating hardening as it depends on the ether content is presented in Fig. 3. Coatings from resins of small ether content achieved a settled hardness after 7-10 days. However, the increase in hardness of coatings from resins of high allyl ether content (above 12 wt%) was initially similar, but after 30 days their hardness was much higher and exhibited a rising tendency. Such drying characteristics may result from hindered oxygen diffusion through the stronger surface crosslinked coating, which occurs as early as the initial step. However, too large an amount of GAE (>16 wt%) is disadvantageous since it causes deterioration of water resistance of the coatings (Table 6).

The quality of resins and coatings depends not only on the amount of GAE but also to a considerable degree on the average functionality of the hydroxylic components. An increase in functionality from 2.28 to 2.32, i.e., a higher content of more branched molecules in the resin, yields coatings with much shorter first

TABLE 1. Feed Composition and Properties of Alkyd Resins Modified by Glycerol Allyl Ether (GAE) (f_{OH} = 2.36)

Resin	GAE, wt%	GAE, mol%[b]	Soybean oil, wt%	Phthalic anhydride, mol%[c]	Substance **2**,[a] wt%	Precondensate acid number, mg KOH/g	Precondensate viscosity, Pa·s	Resin acid number, mg KOH/g	Molecular weight, g/mol
0-1	0	0	59.3	89.1	5.8	20.4	20	43.3	800
1-1	1.9	8.2	56.4	89.6	5.8	21.1	24	38.5	–
2-1	4.0	16.5	53.2	90.4	5.7	21.5	29	40.2	–
3-1	6.4	24.7	50.1	92.7	4.7	25.0	33	37.2	–
4-1	9.2	33.0	46.0	93.5	4.2	26.4	101	42.0	2000
5-1	12.4	40.9	41.5	95.0	3.7	27.2	168	43.3	–
6-1	15.4	48.0	36.5	96.0	3.4	30.0	182	41.0	–
7-1	21.0	57.1	30.4	100.0	0	30.6	Gel	–	–

[a] **2**: 3,3′,4,4′-benzophenonetetracarboxylic dianhydride.
[b] mol% in hydroxylic components.
[c] mol% in acid components.

TABLE 2. Feed Composition and Properties of Alkyd Resins Modified by Glycerol Allyl Ether (GAE) (f_{OH} = 2.32)

Resin	GAE, wt%	GAE, mol%[b]	Soybean oil, wt%	Phthalic anhydride, mol%[c]	Substance **2**,[a] wt%	Precondensate acid number, mg KOH/g	Precondensate viscosity, Pa·s	Resin acid number, mg KOH/g	Molecular weight, g/mol
3-2	6.5	25.2	50.8	91.4	5.2	21.4	41.7	38.0	–
4-2	9.3	33.6	46.9	92.6	4.7	22.1	52.1	39.1	3370
5-2	12.5	42.0	42.1	93.4	4.6	22.3	92.4	41.0	–
6-2	16.2	50.4	36.4	93.9	4.5	22.7	200.0	37.6	4120

[a] **2**: 3,3′,4,4′-benzophenonetetracarboxylic dianhydride.
[b] mol% in hydroxylic components.
[c] mol% in acid components.

TABLE 3. Feed Composition and Properties of Alkyd Resins Modified by Glycerol Allyl Ether (GAE) (f_{OH} = 2.32)

Resin	GAE, wt%	GAE, mol%[b]	Soybean oil, wt%	Phthalic anhydride, mol%[c]	Substance 2,[a] wt%	Precondensate acid number, mg KOH/g	Precondensate viscosity, Pa·s	Resin acid number, mg KOH/g	Molecular weight, g/mol
2-3	4.1	17.2	55.3	90.3	5.5	20.8	14.5	41.4	1600
3-3	6.6	25.8	51.6	90.9	5.4	21.5	17.1	41.0	–
4-3	9.4	34.4	47.5	91.7	5.3	22.7	19.0	39.0	2150
5-3	12.7	43.0	42.7	92.4	5.1	24.2	25.4	42.0	–
6-3	16.5	53.5	37.0	93.0	5.1	19.1	23.0	39.9	3900
7-3	21.2	60.2	30.6	94.9	4.0	25.7	39.0	36.9	–
8-3	26.4	68.8	22.3	94.4	4.8	22.2	104.0	37.4	6000

[a] **2**: 3,3′,4,4′-benzophenonetetracarboxylic dianhydride.
[b] mol% in hydroxylic components.
[c] mol% in acid components.

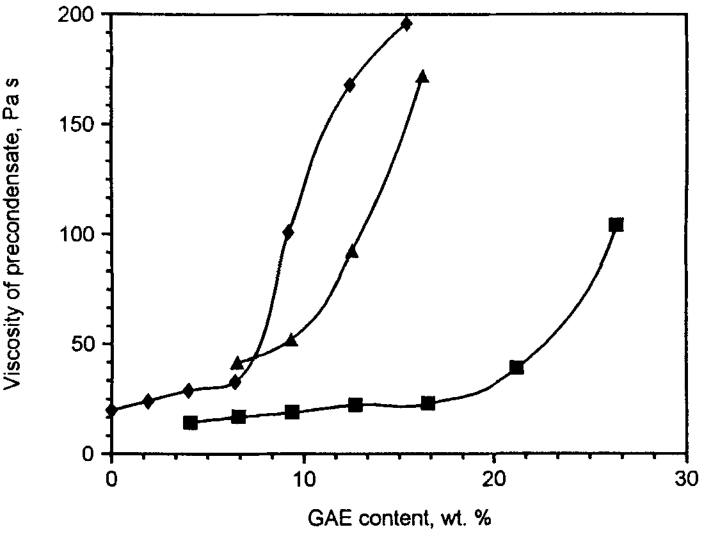


FIG. 1. Viscosity of precondensates depending on the glycerol allyl ether (GAE) content in resins obtained from the pentaerithritol and soybean oil reaction product (**1**) of various functionality (f_{OH}). (♦) f_{OH} = 2.36; (▲) f_{OH} = 2.32; (■) f_{OH} = 2.28.

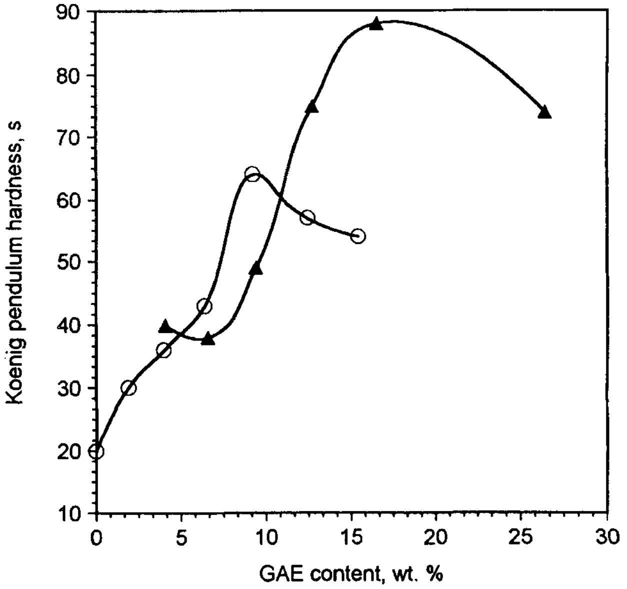


FIG. 2. Hardness of coatings (measured by means of a König pendulum) depending on the share of glycerol allyl ether (GAE) in resins obtained from the pentaerithritol and soybean oil reaction product (**1**) of various functionalities (f_{OH}). (▲) f_{OH} = 2.28; (○) f_{OH} = 2.36.

TABLE 4. Properties of Lacquer Compositions Comprising Alkyd Polyester Resins Modified by Glycerol Allyl Ether (GAE) (f_{OH} = 2.36)

Resin	GAE content, wt%	Hardness K[a] after 3 days, s	Hardness K[a] after 30 days, s	First degree of drying, min	Resistance to water
0-1	0	13 ± 2	20 ± 2	170	+
1-1	1.9	13 ± 3	30 ± 4	130	+
2-1	4.0	13 ± 2	36 ± 3	125	+
3-1	6.4	17 ± 4	34 ± 5	130	+
4-1	9.2	23 ± 3	64 ± 3	130	+
5-1	12.4	20 ± 3	53 ± 5	125	±
6-1	15.4	22 ± 4	54 ± 4	130	±

[a] Hardness measurements were carried out by means of a König pendulum.

TABLE 5. Properties of Lacquer Compositions Comprising Alkyd Polyester Resins Modified by Glycerol Allyl Ester (GAE) (f_{OH} = 2.32)

Resin	GAE content, wt%	Hardness K[a] after 3 days, s	Hardness K[a] after 30 days, s	First degree of drying, min	Resistance to water
3-2	6.5	22 ± 3	55 ± 3	65	+
4-2	9.3	21 ± 2	50 ± 3	70	+
5-2	12.5	22 ± 4	54 ± 5	70	+
6-2	16.2	23 ± 2	70 ± 3	80	+

[a] Hardness measurements were carried out by means of a König pendulum.

TABLE 6. Properties of Lacquer Compositions Comprising Alkyd Polyester Resins Modified by Glycerol Allyl Ether (f_{OH} = 2.28)

Resin	GAE content, wt%	Hardness K[a] after 3 days, s	Hardness K[a] after 30 days, s	First degree of drying, min	Resistance to water
2-3	4.1	15 ± 4	40 ± 3	175	+
3-3	6.6	13 ± 3	38 ± 3	170	+
4-3	9.4	19 ± 2	49 ± 2	110	+
5-3	12.7	26 ± 3	75 ± 5	105	+
6-3	16.5	42 ± 5	88 ± 4	70	+
7-3	21.2	14 ± 2	81 ± 3	120	−
8-3	26.4	18 ± 3	74 ± 3	120	−

[a] Hardness measurements were carried out by means of a König pendulum.

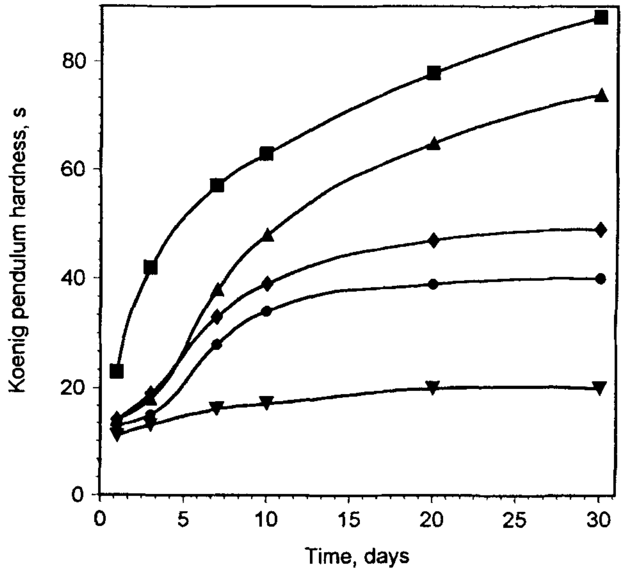


FIG. 3. Hardness of coatings (measured by means of a König pendulum) versus time for various shares of glycerol allyl ether (GAE) in resins (f_{OH} = 2.28). (▲) 26.4 wt% of GAE; (■) 16.5 wt%; (◆) 9.4 wt%; (●) 4.1 wt%; (▼) 0 wt%.

degree drying times. For example, resins 3-3 and 5-3 (f_{OH} = 2.28) reached first degree drying after 170 and 105 minutes, respectively (Table 6), and resins 3-2 and 5-2 (f_{OH} = 2.32) reached first degree drying after 60 and 70 minutes, respectively (Table 5). The final hardness was also reached earlier. A too large functionality leads to longer drying times. Branching does not, however, affect the final hardness of coatings.

TABLE 7. Properties of Lacquer Compositions Made from Resins Containing Glycerol Allyl Ether (GAE) after 3 Months of Storage at 20–23 °C

Resin	GAE content, wt%	Hardness K[a,b] after 3 days, s	Hardness K[a,b] after 30 days, s	First degree of drying, min	Resistance to water
0-1	0	8 ± 1 (13 ± 2)	12 ± 1 (20 ± 2)	230 (180)	–
5-3	12.7	16 ± 2 (26 ± 3)	55 ± 2 (75 ± 3)	116 (105)	±
6-3	16.5	29 ± 2 (42 ± 3)	67 ± 3 (88 ± 5)	110 (70)	+
8-3	26.4	16 ± 2 (18 ± 2)	75 ± 3 (74 ± 2)	130 (120)	–
3-2	6.5	19 ± 3 (22 ± 2)	48 ± 3 (55 ± 3)	86 (65)	+
4-2	9.3	29 (21)	48 (50)	92 (70)	+
6-2	16.2	18 (23)	65 (70)	90 (80)	±

[a] Hardness measurements were carried out by means of a König pendulum.
[b] Values for the composition prior to storage are given in parentheses.

TABLE 8. Effect of Neutralizing Agents on the Properties of the Lacquer Compositions Made from Resins Modified by Glycerol Allyl Ether (GAE)

Resin	GAE content, wt%	Neutralization agent[b]	Hardness K[a] after 1 day, s	Hardness K[a] after 16 days, s
2-3	4.1	TEA	10 ± 2	32 ± 3
2-3	4.1	TEA + ammonia	11 ± 1	47 ± 2
6-3	16.5	TEA + ammonia	11 ± 2	57 ± 4

[a] Hardness measurements were carried out by means of a König pendulum.
[b] TEA: triethylamine.

The resistance to hydrolysis of lacquer compositions containing GAE was studied by comparing the hardness and drying times of coatings applied directly after preparing the composition and from compositions stored at 20-23°C for 3 months (Table 7). The results obtained show that the addition of GAE remarkably improves the composition resistance toward hydrolysis. However, once again, exceeding 15-16 wt% GAE unfavorably affects the stability of compositions.

Attempts carried out to neutralize the studied resins by means of ammonia, which is cheaper and less toxic than amine, showed that complete replacement of the amine is not advantageous, since the neutralized resins could be thinned with water only to 70-60% of their dry mass. Moreover, the quality of the surface worsened. Therefore, an equimolar mixture of triethylamine and ammonia (25% aqueous solution) was used for the neutralization of resins. The results of these studies are presented in Table 8.

Based on our results, we find that resins of $f_{OH} = 2.32$, a precondensate acid number of 20-24 mg KOH/g, containing 10-16 wt% GAE (35-50 mol% hydroxyl components), and 4-5 wt% 2 show the best properties. They exhibit good water solubility (up to 30-40% of the dry mass), good hardness of lacquer coatings (of the order of 50-60 s after 30 days), and a short first degree drying time (in the range of 60-90 minutes). An increase of the GAE content above 16 wt% caused reduced water resistance of the coatings and less stability of the lacquer composition.

CONCLUSIONS

Water-borne polyester alkyd resin susceptible to hydrolysis can easily be modified by allyl ether monomers. Thus, glycerol monoallyl ether (a monomer containing two OH groups) after reaction with the resin introduces unsaturated allyl groups, which are linked to the polymer chain via water-resistant ether bonds. It was shown that the substitution of a part of the fatty acid moieties by a small allyl ether molecule leads to a higher number of reactive double bonds at almost the same molecular weight. This results in a shortening of the drying time and an improvement in the hardness of the coating obtained. The best results were achieved for 10-16 wt% glycerol monoallyl ether in the hydroxylic moieties and for an extent of oligoester branching, i.e., average functionality of the hydroxylic compounds, in the 2.28 to 2.32 range.

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