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Environmentally friendly adhesive composition containing nitrogen

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Abstract: Adhesive compositions used in everyday life, medicine and various industries for gluing many materials must first of all be environmentally friendly, heat-resistant and frost-resistant, have physical and mechanical properties, durability corresponding to operating conditions. For this purpose, oligomers are used as the main component in the system as a binder. Adhesive compositions based on phenol-formaldehyde oligomers modified with compounds of nitrogen-containing amino and amide groups (for example, benzoquinone, benzylamine, benzamide, acetamide, terephthadiamide, urea and oxamide) have been tested and received positive results. To eliminate the disadvantages of the phenol-formaldehyde oligomer (to reduce the number of free monomers, eliminate brittleness, increase heat resistance and adhesion ability), the use of modified co-oligomers as binders in the preparation of an adhesive composition was confirmed for the first time. In the research work, the disadvantages were eliminated by modifying the epoxide-diane oligomer with a nitrogen-containing urea compound, and the basic physico-chemical and physico-mechanical parameters were studied. As a result, the adhesion ability, heat resistance and other indicators of the epoxy-diane oligomer increase. According to the results of spectral analyses, it became known that the modification process is a chemical modification. The copolycondensation reaction at this time was due to functional groups such as epoxide, hydroxide and amide. The modified epoxy-dian oligomer differs from the unmodified epoxy-dian oligomer due to its high adhesive capacity and low build-up during solidification. The main purpose of the research work is to develop an environmentally friendly adhesive composition that is resistant to water, aggressive environments and heat, albeit partially. A new adhesive composition based on a sooligomer has been developed, obtained using urea produced in our republic as a modifier. One of the urgent problems is the growing demand for adhesives with various properties on an industrial scale, respectively, the manufacture of various types of adhesives, including adhesives resistant to heat and aggressive environments.

Keywords: Oligomer, Modification, Cooligomer, Adhesive, Composite, Ecology.

1. İNTRODUCTİON

Adhesive compositions consist mainly of solutions or alloys of oligomers or polymers using other suitable additives as a matrix. The main technological stages of the bonding process consist of the following operations: surface preparation, applying an adhesive composition to the surface, forming or sewing glue.
The components used in the preparation of the adhesive composition must be environmentally and economically beneficial. Of the adhesive compositions, which contain a risk of burning, care should be taken when using them. The adhesive composition is used when gluing two materials of different nature or materials of the same nature in the process of gluing materials of different nature. As a result, the durability of the joints is determined by the adhesive bond strength, that is, adhesion is characterized by the bond strength between two materials of different nature. The strength of adhesive bonds depends not only on the chemical nature of the adhesive composition, but also on the nature and condition of the materials to be bonded.

Adhesives can be both organic and inorganic. By itself, the organic adhesive composition can be of two types: natural and artificial. Natural adhesives include starch and casein. Natural macromolecular compounds are used as the main raw material for the manufacture of artificial adhesives. Synthetic adhesives, on the other hand, are compounds synthesized by chemical methods such as polycondensation, copolycondensation, polymerization, copolymerization or polymeranalogical transformation reactions [1-10].

Such compositions are called inorganic adhesives if the matrix of the adhesive composition is an inorganic compound (cement, gypsum, liquid glass, etc.).Adhesives based on epoxy-diane oligomer have high adhesion rates compared to adhesives based on other oligomers.

Considering that the number of epoxy groups in the polymer obtained on the basis of diphenylolpropane, epichlorohydrin and urea is 15.4-16.5%, an adhesive composition is made from it. Polyethylene polyamine or nitrogenous organic compounds are mainly used as fasteners for fixing the finished modified composition based on epoxy-diane cooligomer. The amount of its fasteners added to the adhesive composition also affects the heat resistance of the adhesive. During the curing process, the adhesive composition changes from a viscous state to a solid one, capable of wetting the surfaces to be bonded well or penetrating into the pores of the surfaces.

Epoxydian oligomer-based adhesive compositions have the following advantages compared to existing oligomers belonging, for example, amine-aldehyde, phenol-formaldehyde, resorcin-formaldehyde-based adhesive compositions:

- ability to adhesive materials of different nature;
- eliminates corrosion;
- the hardening process is hardened at a temperature not higher;
- possible to carry out the solidification process at room temperature.

Epoxy resin based adhesives can be used in various industries such as construction, automotive, aerospace and electronics due to their versatile and customizable properties. Ongoing research in this direction is aimed at optimizing the composition and main indicators of functioning adhesives based on cooligomers and lead to advances in the technology of preparation of adhesive composition. The importance of preparing an adhesive composition based on the functional epoxide-diane cooligomer is due to the fact that, in modification with urea, various indicators of the epoxide-diane cooligomer are improved, such as elasticity, strength and adhesion strength, which makes the adhesive suitable for specific applications.

To improve the fire resistance of epoxy-Diane oligomers, modifiers such as graphene, phosphorus, boron compounds or similar suitable fillers are used. The number of modifiers introduced into the matrix based on the epoxide-diane cooligomer is very important.because otherwise it can lead to undesirable thermal and mechanical properties.

2.EXPERIMENTAL DETAIL

Modification of the epoxide-diane oligomer with urea was carried out in a laboratory reactor and the adhesive ability of the resulting co-oligomer of a new composition was studied. In a 250 ml flask equipped with a blender, a reverse cooler, a drip lock and a thermometer, 5 g of dry sodium hydroxide is dissolved in 43.3 ml of distilled water. 20 g of diphenylolpropane is added to the prepared alkali solution, stirring. After diphenylolpropane is completely dissolved, epichlorohydrin is gradually added to the mixture in an amount calculated from the drip lock. The mixture is heated in a water bath at 70 °C for 45 minutes, after 45 minutes a second portion of an alkali solution is added to the flask (obtained by dissolving 2 grams of NaOH in 6 ml of distilled water) and the mixture is heated at 82 °C for 30 minutes. Then a third part of the alkali solution (consisting of 0.73 g of NaOH and 25 ml of distilled water) is added to the flask and the calculated amount of urea is gradually added. The temperature is brought to 95 °C and heating at this temperature continues for an hour. The resulting cooligomer is passed through a flask into a crucible, washed several times with water until a neutral reaction (the water temperature should be 50 °C). It is then dried in a vacuum drying cabinet until it reaches a constant weight. Dried epoxy-diane cooligomer of light yellow color. Thus, the solubility of the obtained cooligomer of a new composition in organic solvents, the number of characteristic epoxy groups, the mass of the molecule, and the characteristic viscosity are determined.

To prepare an adhesive composition based on a relatively environmentally friendly new epoxy-diane cooligomer, MgO (2-10%) was used as a filler, and dibutyl phthalate (1-3%) was used as a plasticizer. First, a 50% solution of the epoxide-diane cooligomer is prepared in an organic solvent, then these components are added. The physico-mechanical and operational parameters of the finished adhesive composition have been studied [11-16].

A physico-chemical, physico-mechanical and spectral analysis of the cooligomer and the adhesive composition based on the sooligomer was carried out.

Table 1. Physico-chemical and physico-mechanical parameters of the modified epoxide-diane cooligomer and the unmodified epoxide-diane oligomer

№	The name of the indicator	Indicators	
		Epoxy-diane oligomer	Epoxy-diane cooligomer
1	**2**	**3**	**4**
1	Amount of nitrogen, %	-	7.6 - 8.2
2	Number of epoxy groups, %	20 - 22	15.4 - 16.5
3	Number of hydroxyl groups, %	16.8 - 17.4	12.6 - 14.2
4	Molecular weight	350 - 400	2068 - 3102
5	Density, kg/m^3	1042.4	1080 - 1100
6	Gelatinization time at 160°C, sec.	159 - 165	130 - 135
7	Softening temperature, °C	60 - 65	68 - 72
1	**2**	**3**	**4**
8	Degree of solidification at 150 ° C for 5 hours, %	92 - 94	98 - 99
9	Heat resistance accor-ding to Vica, °C	120 - 140	150 - 160
10	Adhesive strength, MPa	18 - 20	30 - 34
11	The viscosity of a 50% solution is in the VZ-4 device, sec.	45 - 50	55 - 60

A number of physico-chemical and physico-mechanical parameters of the urea-modified epoxide-diane cooligomer have been studied and it has been established that the physico-chemical and physico-mechanical parameters of the modified epoxide-diane cooligomer are better than the physico-chemical and physico-mechanical parameters of the unmodified epoxide-diane oligomer.

The IR spectral analysis of the unmodified epoxide-diane oligomer, the modified epoxide-diane cooligomer and the adhesive composition based on

the cooligomer was studied (Fig.1-3). The IR spectrum was obtained using the Thermo scientific infrared spectrometer.

Here 3404,35 cm^{-1} indicates the valence vibration of the OH group in the absorption band, 3067,82 cm^{-1} indicates the valence vibration of the CH and epoxy group in the absorption band, 2956,82 cm^{-1} indicates the valence vibration of the CH_3 group in the absorption band, 1455,16 cm^{-1} indicates the deformation vibration of the CH_3 group in and in its strip indicates the presence of deformation vibration of epoxy groups.

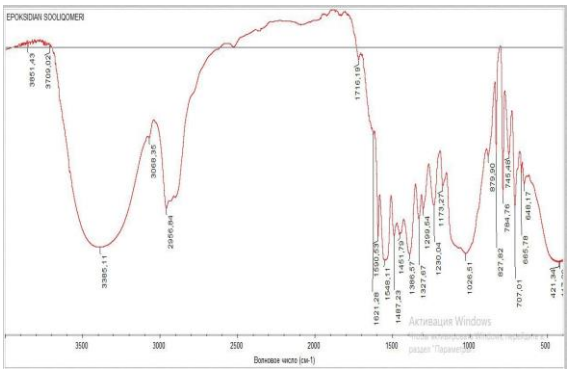

Fig. 1. IR spectrum of modified epoxy-diane cooligomer.

Looking at the peaks in the IR spectrum of the urea-modified epoxy-diane cooligomer, we observe the presence of CO, NH, OH, CH, CH_2, CH_3 and the epoxy group.

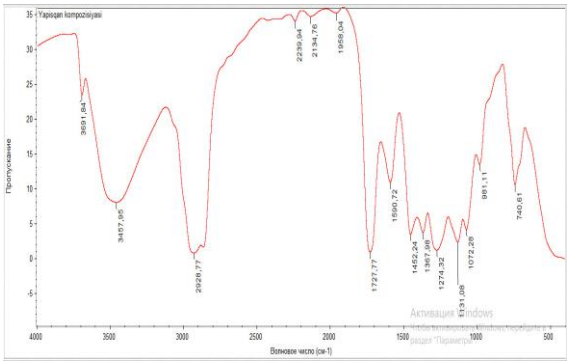

Fig. 2. IR spectrum of modified epoxy-diane cooligomer.

In this IR spectrum, 3385,11 cm^{-1} absorption band reflects the valence vibration of the NH and OH groups in the coligomer, 2956,84 cm^{-1} absorption band reflects the valence vibration of the CH Group, 1548,11 cm^{-1} the deformation vibration of the CO and NH groups in the absorption band, 1327,67 CH_2 and deformation vibration of the CH groups 1230,04 cm^{-1} indicates deformation vibration, 827.82-707.01 cm^{-1} absorption band indicates deformation vibration of epoxy group.

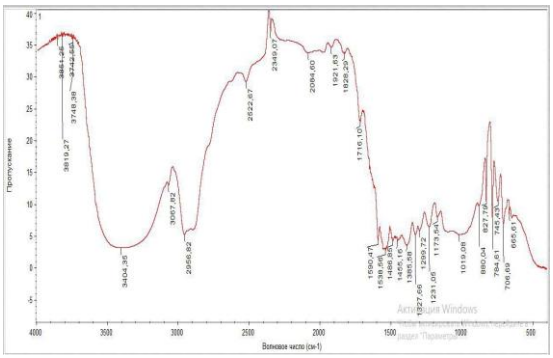

Fig. 3. Epoxy-diane coligomer based adhesive composition modified with urea

Looking at the IR spectrum of the adhesive composition, we observe that there are groups of OH, NH, CO, CH and benzene. Here, 3457.95 cm^{-1} absorption band indicates valence vibration of the OH and NH group, 2928.77 cm^{-1} absorption band indicates valence vibration of the CH group, 1727, 77 cm^{-1} absorption band indicates valence vibration of the CO group, 1590.72 cm^{-1} absorption band indicates deformation vibration of the NH group, 1274.32 cm^{-1} absorption band indicates deformation vibration of the CO group.

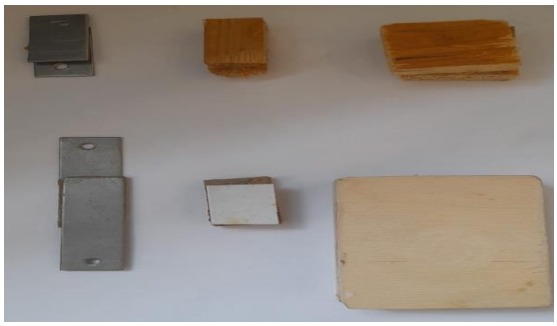

Fig. 4. Adhesive substrates of different nature

The adhesive composition based on a carbamide-modified epoxy-diane cooligomer in Figure 4 was used to glue substrates of the same nature. The degree of adhesive adhesion is determined here by gluing iron to iron, wood to a board, and ceramics to ceramics. The adhesive composition is designed for gluing materials of the same nature together, as well as for gluing materials of different nature.

3. CONCLUSION

An unmodified epoxide-diane oligomer was synthesized and some of its parameters were studied. The epoxy-diane oligomer has been modified with urea and the physico-mechanical, physico-chemical parameters of the modified epoxy-diane cooligomer have been established. It was found that in the epoxy-diane cooligomer with the function of urea, urea eliminates the disadvantages of the epoxy oligomer.
It was found that the main indicators of the modified cooligomer-based adhesive composition are 1.5 times higher than the indicators of the unmodified oligomer-based adhesive composition. When modifying the epoxide-diane oligomer with urea, an increase in functionality and an improvement in its properties were revealed, which was reflected in the methods of IR spectral analysis. An adhesive composition was developed with the addition of filler and plasticizer to a modified epoxy-diane cooligomer. The indicators of the adhesive composition have been established. The adhesive composition is made by applying different substrates and gluing different substrates to each other. It has been established that the adhesive composition glues wood, iron, ceramic and other substrates to each other.

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