Study of thermal state, water absorption and strength properties of composite polymer-polymer binders

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Abstract. In this article, in order to improve the heat-water resistance and strength properties, the modification of urea-formal- dehyde polymeric materials is considered by introducing melamine and acrylonitic resins and reactive compounds into their composition. Shown by identifying the optimal ratios of urea-formaldehyde, melamine and acrylonitrile resins and the corresponding reactive compounds in increasing the heat-water resistance and shear strength of adhesive joints basedon them, replacing expensive phenol-formaldehyde resins used in gluing basalt fibrous masses in the production of build-ing heat-insulating sheet materials.

1 Introduction

Today, there is a great demand in the world for building materials, including heat-insulating sheet materials, where their preparation requires non-deficient polymer binders with high physicochemical, heat-resistant and other operational properties [1,2]. In this regard, one of the important tasks in this direction is the development of composite polymer binders and adhesives based on them from local and secondary raw materials with high heat- and water-resistant properties for the production of building heat-insulating sheet materials and other industries. As it is known [3-14] adhesives are individual substances or a mixture of organic, organoelement or inorganic compounds that have good adhesion, cohesive strength, sufficient plasticity and durability under application conditions and are able to be approved with the formation of strong adhesive compounds.

In the world, scientific research on the creation of efficient and cheap building materials, including composite polymer binders and adhesives based on them from local raw materials and industrial waste with high heat and water-resistant properties, used in the production of building sheet insulation and other materials, is of great importance. Therefore, the development and improvement of physico-chemical properties and improving the quality of composite polymer binders and adhesives based on them with high heat and water-resistant characteristics, as well as improving the technology of their production, is of particular importance today [15-24].

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Research is being carried out in the Republic of Uzbekistan and certain results have been obtained in the development of composite polymer binders and adhesives based on them from local raw materials and industrial waste. However, these developed composite polymer binders do not sufficiently meet the requirements of the production of building heat-insulating sheet materials, especially on the basis of basalt fibrous materials produced in Uzmetkombinat JSC.In this aspect, the development of effective import-substituting composite polymer binders and adhesives based on them from local and secondary raw materials and technologies for their production, especially heat- and water-resistant, is an urgent problem[25-32].

The aim of the study is to investigate the possibility of increasing the heat and water resistance of urea-formaldehyde polymer binders and shear strength of adhesive compounds based on them by chemical modification of their introduction into the composition of melamine and acrylonitrile resin, as well as reactive compounds.

2 Research methods

The objects are urea-formaldehyde, melamine and acrylonitrile resins, caustic sodium, zinc chloride, urotropin, ammonium chloride, and compositions based on them, as well as glue.

Physico-mechanical heat resistance, water absorption, shear strength of adhesive compounds, properties of the developed composite polymer binders were determined by generally known methods [4-8], allowed in the CIS countries.

Results and discussion of the study. To conduct research in the field of chemical modification in order to increase the heat and water resistance of a urea-formaldehyde polymer binder, we selected acrylonitrile resin having high heat resistance, and chalk resin having high water resistance, as well as reactive compounds. In accordance with the concepts being developed, it was necessary to find out the effect of melamine and polyacrylonitrile on the process of structure formation of the composition and, ultimately, on the heat-water resistance of the urea-formaldehyde polymer binder.

Further, in order to identify an effective hardener, the heat resistance of the polymer-polymer compositions being developed, cured with various reactive compounds, was investigated using the example of urea-formaldehyde and polyacrylonitrile compositions. The heat resistance of polymer compositions was investigated by the method [1,2].

3 Results and discussion

Table 1 shows the values of the heat resistance of polymer-polymer binders- adhesives depending on the ratio of urea-formaldehyde resin (MFS) and polyacrylonitrile (PAN) cured with various reaction compounds.

As can be seen from Table 4.4, the heat resistance of composite binders increases monotonically with an increase in the content of polyacrylonitrile in the composition of urea-formaldehyde resin. High values of heat resistance of polymer-polymer compositions are observed in the following ratios of urea-formaldehyde resin and polyacrylo-nitrile from a ratio of 50:50 to a ratio of 10:90. At a ratio of 50:50, the composition has 250°C- 256°C; at 40:60 - 260°C- 275°C; at 30:70 –286°C-292°C; at 20:80 - 292°C-306°C; at 10:90 -300 With 326°C, which fully meets the requirements of the production of thermal insulation sheet materials based on basalt fibrous materials.

Table 1. Heat resistance of the considered composite polymer-polymer binders depending on the ratio of urea-formaldehyde resin (MFS) and polyacrylonitrile at various hardeners.

№	**The ratio of MFS and PAN, mass.h**	**90:10**	**80:20**	**70:30**	**60:40**	**50:50**	**40:60**	**30:70**	**20:80**	**10:90**
	Hardeners, mass.h	**Heat resistance of the material**								
1	Caustic sodium (40%) 0.3-0.7	120	150	180	210	250	270	290	300	320
2	Zinc chloride (50%) 1,62	125	156	190	220	255	275	292	306	326
3	Urotropin 7.7	118	148	186	208	248	268	286	292	300
4	Ammonium chloride 0.025-0.1	122	152	186	212	256	272	290	302	322

At the same time, it can be noted that the heat resistance of the polymer-polymer composite binders-adhesives cured with various reactive compounds by catalysts, with minor changes, is basically within the same limits. However, it should be noted that they differ significantly in cost. So, the price of 1 kg of caustic sodium costs 19.500 soums or 150 rubles (flagma.uz), zinc chloride-80.000 soums or 400 rubles (flagma.uz), urotropin- 40.000 soums or 200 rubles (flagma.uz), ammonium chloride-20.000 soums or 100 rubles (flagma.uz) on 10.07.2022. From an economic point of view, for the production of polyacrylonitrile-formaldehyde composite polymer binders, adhesives, we further used ammonium chloride as a catalyst hardener during curing. (temperature 120°C and time 2 hours).

The study found that the best and heat-resistant properties, as well as low water absorption in composite materials based on mochovino-formaldigin resin and polyacrylonitrile are observed at their ratio of 50 to 50 and 60 to 40 wt.h, as well as composite binders based on mochovino-formaldigin resin and melamine resin in 100 on 6 ratios, the indirect ones are named MFS-PAN-1, MFS-PAN-2 and MFS-MS-1. In this regard, we further investigated the heat resistance of the developed composite materials of the brands MFS-PAN-1, MFS-PAN-2, MFS-MS-1 and the well-known glue K-153, produced in Russia, under the prolonged action of elevated temperature. Table 2 shows comparative data on thermal aging of composite adhesive compounds based on K-153 glue and the composite polymer-polymer bonding adhesives MFS-PAN-1, MFS-PAN-2 and MFS-MS-1 that we are developing.

Table 2. Comparative data on thermal aging of adhesive compounds in composite polymer-polymer adhesives at operating temperatures up to 60°C.

Brand of glue	**Aging conditions**		**Destructive shear MPa**					
	°C	**Sell flax-news, h**	**In the initial state**			**After aging**		
			- 60°C	**20°C**	**60°C**	**- 60°C**	**20°C**	**60°C**
K-153	60	500	9.1	9.7	9.0	9.5	10.5	8.9
MFS-MS-1	60	500	17.4	18.0	17.8	18.4	19.2	16.2
MFS-PAN-1	60	500	29	31	30	29	32	30
MFS-PAN -2	60	500	28	30	29	30	31	28

When considering composite adhesives, as can be seen from Table 1, at a temperature of 600C for 500 hours, the best results were shown by our composite polymer-polymer bonding adhesives of the MFS-PAN-1 and MFS-PAN-2 brands.

Thus, the shear strength of K-153 in the initial state is within 9.7 MPa, and the MAS-PAN-1 adhesive is 18.0 MP. Composite polymers-polymer adhesives MFS-PAN-1 and MFS-PAN-2 are in the range of 31.0 MPa and 30.0 MPa. Before and after the test, within the minus degree - 60°C and plus +60°C, the heat resistance decreases slightly.

For example, when testing +60°C, the heat resistance of K-153 glue is 8.9 MPa, and MFS-PAN-1 is 16.2 MPa. The shear strength of the developed composite polymer-polymer adhesives MFS-PAN-1 and MFS-PAN-2 is in the range of 30 MPa and 28 MPa, respectively. This may well meet the requirements of the production of heat-insulating basalt sheet materials in the conditions of JSC "Uzmetkombinat".

It should be noted that with the prolonged effect of elevated temperature on the adhesive joints, a change in strength occurs due to thermal or thermal oxidative degradation, or due to the action of thermal stresses due to the difference in the coefficients of linear expansion of the materials to be bonded and the glue. The latter circumstance is mostly decisive when using adhesive joints in low temperature conditions.

Temperature or sudden temperature drop. If the materials to be bonded dry out under the action of temperature and deform at the same time, then stresses (humidity) also arise, which can be more destructive than thermal ones. Therefore, it is very important to find out the predominant mechanism of aging.

The general patterns that manifest themselves during thermal aging of adhesive joints are mainly reduced to the following.

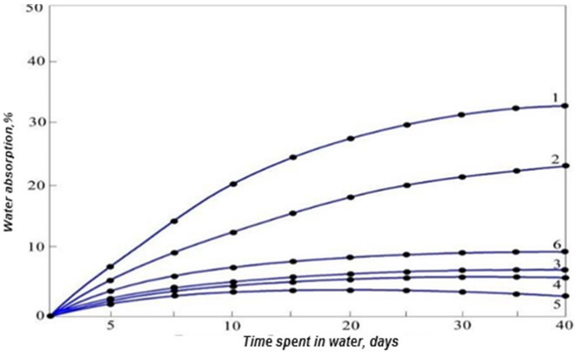
Rigid strongly crosslinked polymers are most resistant to thermo acid, but in the process of thermal aging they experience the greatest overvoltages, which will lead to a significant decrease in strength with a small loss of mass. More rare or elastic bonds contribute to the relaxation of overstrain.

The same effect is achieved when applying elastic polymer primers under hard glue.

With prolonged exposure to elevated (and sometimes lowered) temperatures, the nature of the surface of the materials to be bonded may change.

The water absorption of the polymer-polymer compositions being developed was also investigated depending on the time of separation in water.

Figure 1 shows the results of studies of the dependence of the water absorption of the developed modified polymer-polymer composite binders-adhesives on the duration of the long stay of their samples in water for 40 days.



The ratio of urea - formaldehyde resin to polyacrylonitrile is as follows: 1-90:10; 2-70:30; 3-50:50; 4-40:60;5-10:90; 6-100 wt.h WT:6 wt.h of melamine

Fig.1. Dependence of water absorption of polymer-polymer compositions on the residence time of the sample in water at different ratios of urea-formaldehyde resin and polyacrylonitrile.

As can be seen from Figure 1, all the samples considered have 90:10; 70:30; 50:50; 30:70 and 10:90 it is observed that water absorption increases with an increase in the holding time of samples in water. At the same time, the lowest water absorption is observed in samples with a ratio of MFS and PAN 50:50; 40:60 and 10:90.

As can be seen from the curves 3,4,5,6 of Figure 1, the degree of water absorption of samples increases in all cases and within 30 days. With further stay in the water, the water absorption of the samples slows down significantly and practically remains at this level for up to 40 days of stay in the water, that is, there is no increase in water absorption.

For comparison, the water resistance of the composition was studied in parallel at the ratio of WT and PAN at numbers 1 and 2. As can be seen from curves 1 and 2, the water absorption of these samples differs sharply from the modified samples and is in the region of 30% and 22%, respectively.

The adhesive properties of polymer compositions MAS-PAN-1, MS-PUNK-2, MS-MS-1 and glue K-153 were investigated.

Table 3 shows the changes in shear strength of adhesive joints on the specified poly-dimensional composite binders with glass after holding in water for 30 days at a normal temperature of 20 -22°C in storage conditions for 5 months and on an open site for 5 months at a test temperature of 20-22°C.

Table 3. Change in shear strength of glass adhesive joints after exposure for 30 days in water, in a warehouse.

Type and brands of polymer binder-glue	**Glued materials**	**Reduction of shear strength after exposure at temperature 20-25°C**		
		In water for 30 days	**In warehouse conditions for 5 months**	**On an open site for 5 months**
K-153 Russian production	Glass	20	5	5
MFS-PAN-1	Glass	10	5	5
MFS-PAN -2	Glass	6	4	3
MFS-MS-1	Glass	5	3	2

As can be seen from Table 3, the viciousness of the K-153-based adhesive compounds and modified composite polymer adhesives MFS-PAN-1, MFS-PAN-2 and MFS-MS-1 at normal temperatures of 20-22°C decreases by 20% and 10% during 30 days in water, 6% and 5% respectively. It can be seen from this that the shear strengths of the MAS-PAN-1, MFS-PAN-2 and MS-MS-1 compositions will be better preserved in water compared to K-153, and in storage conditions and in the open area at the MFS-PAN-1 and MFS-PAN-2 com positions, the strength at the shear is slightly better than glue K-153.

4 Conclusion

The influence of various reactive compounds (caustic sodium, zinc chloride, urotropin and ammonium chloride) on the curing process of composite binders based on urea-formaldehyde resin and polyacrylo-nitrile at their various ratios was investigated. At the same time, it is established that the heat resistance of polymer-polymer composite binders-adhesives rejected by these hardeners, with minor changes, is mainly within the same limits. Taking into account the economic point of view of the building, ammonium chloride was chosen as a hardener for further research.

It was found that with the prolonged effect of the above temperature on the adhesive joints on based on polymer binders K-153, MFS-MS-1, MFS-PAN-1, MFS-PAN-2 The best results were shown by composite polymer-polymer bonding adhesives based on MFS-PAN-1 and MFS-PAN-2 The water resistance of the polymer binders under consideration based on polymer the MFS-PAN binder with their various correlations. At the same time, the lowest water absorption is observed in samples of polymer composite binders with their ratio of MFS and PAN 50:50, 40:60 and 10:90.

It has been established that the media of the studied composite polymer adhesive compounds with glass at exposure of 30 knocks in water of the shear strength of the composition based on MFS-PAN-1 and MFS-PAN-2 and MFS-MS-1 will be better preserved in water by tearing off the glue K-153, and in storage conditions and in the open However, the compositions MFS-PAN-2 and MFS-MS-1 have somewhat better shear strength than K-153 adhesives.

Thus, experimental research has established that among the composite polymer-polymer binders of the brand of adhesives based on modified urea-formaldehyde resins and K-153 adhesives, the best heat and water resistance and shear strength in water developed composite binder adhesives of the MFS-PAN-2, MFS-PAN-1 and MFS-MS-1.

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