Nat. Volatiles & Essent. Oils, 2021; 8(4): 5635-5643

# Development Of An Effective Import-Substituting Composite Polymer Adhesive And Technology For Their Production

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**Abstract:** Analysis of the current state of polymer adhesives and their application in various industries, including for affixing the markings of excise-branded alcoholic beverages; based on the results of the study of the type, structure, composition and nature of organic and inorganic ingredients in local and secondary raw materials for the production of composite polymer adhesives for use in the production of import-substituting alcoholic beverages. to study the influence of the nature, structure, composition and quantity of polymer binders and organomineral ingredients from local and secondary raw materials on the formation of the physicochemical and strength properties of the resulting composite polymer adhesives; development of optimal composition of composite polymer adhesive adhesives based on polymer binders and organomineral ingredients from local and secondary raw materials for use in the production of alcoholic beverages and other industries; opinions and comments on the development of technology and production of experimental batches of composite polymer adhesives from organomineral ingredients based on local and secondary raw materials, development of scientific and methodological principles of production technology and calculation of feasibility.

**Keywords:** polymer, organomineral, adhesives, secondary raw materials, organomineral.

## Introduction

Today, the demand for polymer adhesives used for bonding various materials in industrial enterprises, construction and furniture industry in the world is growing day by day. In this regard, the affixing of paper labels on metal, glass, plastic, cardboard and other containers, the creation of composite polymer adhesives with high adhesion-cohesive properties for affixing excise stamps of alcoholic beverages are of particular importance for the food industry.

In the world, scientific research on the creation of effective composite polymer adhesives with cheap and high physicochemical properties on the basis of industrial waste for affixing paper labels and other introductory warning signs, affixes of excise stamps of alcoholic beverages is of great importance. In this regard, the creation and improvement of quality of effective composite polymer adhesives with high adhesion-cohesive properties, improvement of physical and chemical properties, as well as improvement of technology for their production are of particular importance today.

Research work on the production of glue on the basis of local raw materials and industrial waste is being carried out in the country, and certain results are being achieved. The fourth chapter of the Action Strategy for the further development of the Republic of Uzbekistan identifies important tasks on "... the use of effective mechanisms, scientific and innovative developments to stimulate research and innovation ...". In this regard, the development of effective composite polymer adhesives and their production technology from organomineral ingredients based on local raw materials and industrial wastes, research aimed at obtaining import-substituting composite polymer adhesives with high physicochemical properties, strength of adhesives obtained at different temperatures, obtained composite polymer adhesives scientific research on methods of determining shelf life and swelling is of great importance.

**The degree to which the problem has been studied**

The following scientists have contributed to the creation and development of composite polymer and adhesive materials: Tadashi Ashida, Rahman Kamali, S.Huicmand, R.Morgen, A.D. Amore, D.Jully, G.Akovali, N.S. Enikolopov, S.N. Jurkov, V.V. Korshak, A.S. Wolfson, A.A. Berlin, M.S. Akutin, Yu.S. Lipatov, E.F. Oleynik, F.Mettyuz, G.S. Golovkin, J.X. Xalikov, M.A. Askarov, S.S. Negmatov, S.Sh. Rashidova, G.Raxmonberdiev, A.T. Djalilov, T.M. Boboev, X.I. Akbarov, A.Umarov, N.S. Abed, A.X. Yusupbekovs, A.Kumar, H.J. on the development of technology for obtaining composite polymeric materials and adhesives based on them. Deppe, K.Ernat, H.Sane, A.A. Moslemi, H.A. Miller, V.A. Belogo, A.I. Sviredenok, M.I. Petrocoven, E.I. Karaseev, S.A. Ugryumov, V.N. Volyunskiy, G.I. Schwartzman, V.V. Gluxix, G.A. Golubitskaya, V.M. Kurlyumov, V.G. Savkin, A.V. Struk, V.N. Solomko, R.G. Maxkamov, A.Salimsakov, A.A. Riskulov, F.A. Magrupov, R.S. Sayfutdinov, Z.N. Muxitdinov, K.S. Negmatova, B.X. Tulyaganov, A.S. Dedicated to the works of Ibadullaev and others.

**MAIN PART**

The properties of the selected chemical fibers were studied in order to study the effect of binders for the formation of composite polymer adhesives. The structure, physicochemical and strength properties of the produced composite polymer adhesives, as well as their technological and physico-mechanical, adhesion-cohesive properties are determined by the initial properties of the organomineral ingredients used in the production of adhesives. To do this, first of all, the composition, structure, physicochemical and strength properties of the organomineral ingredients and chemical fibers that make up the components of the glue were studied.

The physical and mechanical properties of the following components of the composition for the selection of polymer binders for the development of composite polymer adhesives were studied: nitron, kapron, lavsan, acetate, triacetate and viscose fibers.

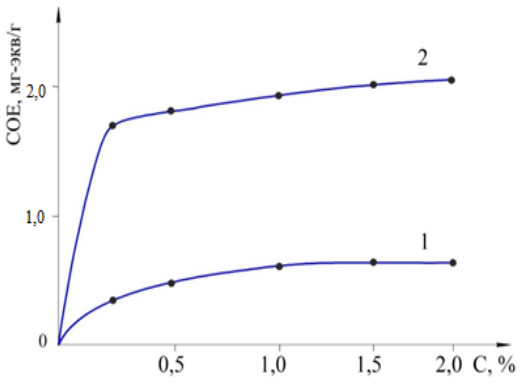
Studies have shown that nitron fiber is the cheapest, the thinnest fiber and has the best physicochemical properties. Therefore, nitron fiber (polyacrylonitrile - PAN) was subsequently selected as a

polymer binder for the production and production of import-substituting composite polymer adhesives for affixing excise-branded alcoholic beverage labels.

It should be noted that local adhesives made from unmodified polyacrylonitrile 70% and 20% alkali solutions do not meet the requirements for the production of alcoholic beverages. This, in turn, requires improving the adhesion and cohesion properties of adhesives derived from polyacrylonitrile, which will require modification of the polyacrylonitrile fiber.

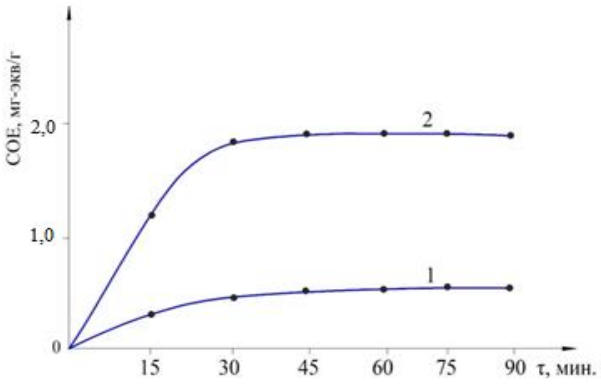
In this regard, first of all, the modification of polyacrylonitrile (PAN) fiber with an aqueous solution of sulfuric acid - H2SO4 was studied.

From the obtained data, it can be seen (Fig. 1) that an increase in the concentration of sulfuric acid from 0.25 to 0.5% leads to a sudden increase in the static exchange capacity (SAS) of the PAN fiber. Subsequent increases in the concentration of sulfuric acid in solution from 0.5 to 2.0% did not lead to significant changes in the static exchange capacity of the modified nitron fiber. The kinetics of hydrolysis of nitron fiber with 0.5% sulfuric acid solution at temperatures of 353 K and 373 K were studied (Figures 1, 2). From the data obtained, it can be seen that increasing the hydrolysis process to 30 minutes leads to an increase in the SAS of the nitron fiber.




1- 353 К; 2-373 К;

**Figure 1. Influence of sulfuric acid concentration on nitron fiber modification at different temperatures**




1- 353 К; 2-373 К;

**Figure 2. Kinetics of the process of hydrolysis of nitron fiber with sulfuric acid solution at different temperatures**

Over time, a significant increase in fiber SAS during hydrolysis occurred at 373 K. As a result of hydrolysis for 30–35 min at 373 K, the SAS of PAN fiber was 2.1–2.3 mg-eq / g. When the hydrolysis process was continued again, the SAS of the PAN fiber was slightly reduced, which can be explained by the hydrolysis of the urea groups to the carboxyl groups.

Thus, to modify nitron fiber with sulfuric acid, it is recommended to process it at an acid concentration of 0.5%, a temperature of 373 K, and a time of 30-35 minutes.

In addition, the effect of sodium hypochlorite concentration on the amount of amino group formed in nitron fiber was studied (Fig. 3). The reaction was performed at 353 K for 20 min by varying the sodium hypochlorite concentration from 1.0 to 10 g / l. The data obtained show that an equilibrium was reached when the concentration of sodium hypochlorite was 4.2 g / l and an amino group of 1.75–1.85 mg-eq / g was formed in the fiber. The kinetics of the oxidation process of nitron hydrolyzed at 353 K with 4.2 g / l sodium hypochlorite solution was studied (Fig. 4). The results show that as the duration of the oxidation process increases, the amount of amino groups increases and reaches an equilibrium value within 15-20 minutes.

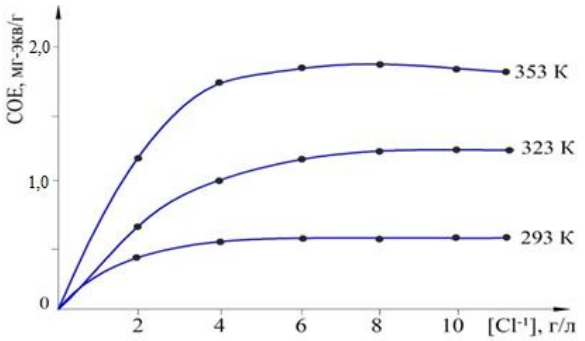



**Figure 3. Effect of sodium hypochlorite (NaClO) concentration on nitron fiber modification**

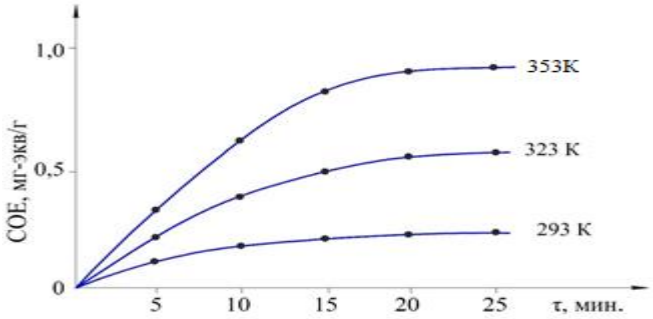
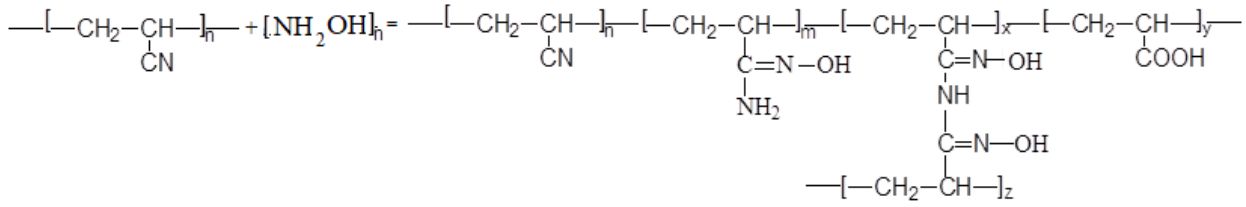



**Figure 4. Kinetics of oxidation of hydrolyzed nitron fiber with sodium hypochlorite (NaClO) solution**

The results of the study show that the optimal conditions for the modification of nitron fiber with sodium hypochlorite in order to obtain nitron fiber, which preserves amino groups:

Different concentrations of hydroxylamine solutions were used as the modifying agent. The reaction mechanism of this process is as follows:

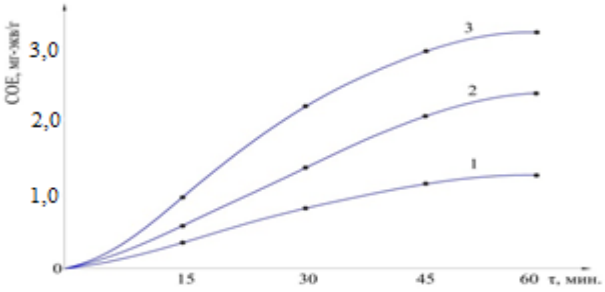


When polyacrylonitrile fibers react with hydroxylamine, functional groups of polyacrylamidoxime are formed. However, amidoxylation of nitron fiber at 90 0S is performed to obtain anion-exchange and complex-forming fibrous materials. At the same time, the properties of the fiber, ie chemical resistance and mechanical strength, deteriorate. This makes it easier to develop an adhesive composition under the influence of alkali.

The kinetics of the nitron fiber modification process were studied for solutions with hydrochloric acid concentrations of 6 g / l, 9 g / l, 12 g / l, 15 g / l, and 18 g / l at the boiling point of a 5% solution of DMF (Figure 5). .

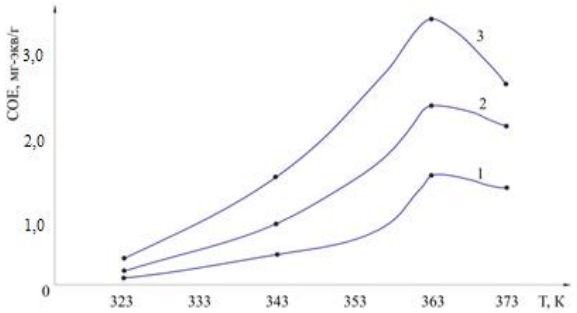
It should be noted that when hydroxylamine is exposed to PAN fibers, first amidoxime groups are formed, which are then converted to urea groups.

The effect of temperature on SAS on NSl of the obtained samples was studied. As shown in Figure 6, initially the modification process takes place on the surface of the fiber, and then as the modification duration increases and the concentration of hydroxylamine in the solution increases, the polymer fiber undergoes a profound change, leading to an increase in SAS value (Figure 6).




1, 2, 3 - 6 g / l, respectively; 9 g / l; 12 g / l ;

**Figure 5. Kinetics of the process of modification of nitron fibers with hydroxylamine solutions of different concentrations**




1, 2, 3 - 6 g / l, respectively; 9 g / l; 12 g / l ;

**Figure 6. Modification of SAS depending on the reaction temperature of the modified nitron fiber at the following concentrations of hydroxylamine**

The best results were obtained when the modification process was carried out when the pH of the initial solution was 6.5-7.0. Adhesion, cohesion and physicochemical properties, as well as adhesion and adhesion strength of polymers depend to some extent on their structure, chemical composition and molecular weight.

Considering the bonds in the structure and chemical structure of polymers with adhesive properties, a number of examples can be given to substantiate the effect of the nature of functional groups on the adhesion and cohesion properties of monomers and polymer compounds.

Figure 7 shows the IR spectrum of Nitron fiber-based glue.

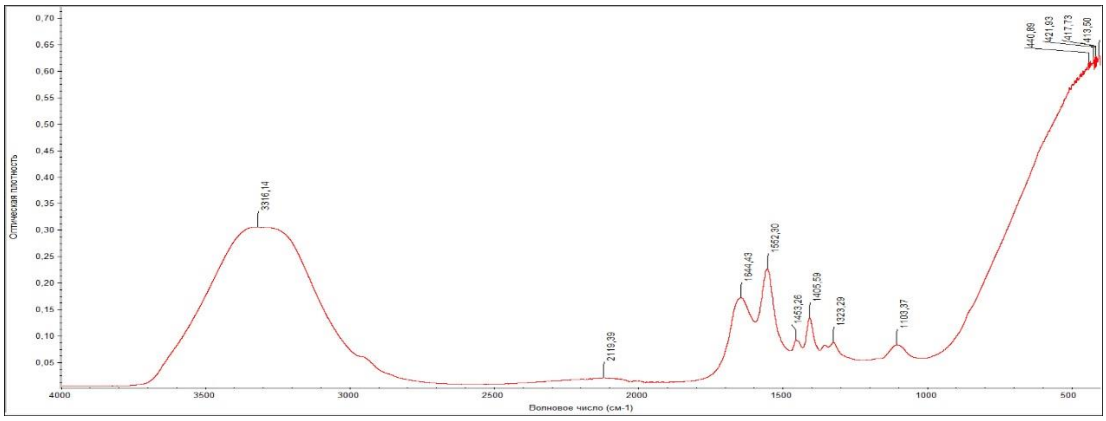


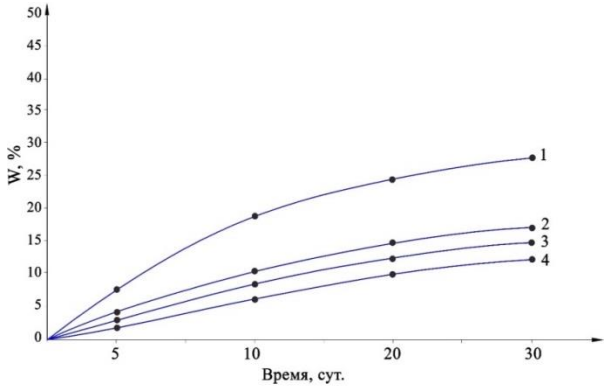

**Figure 7 IR spectrum of glue based on "Nitron" fiber**

The flattening of the given absorption lines is based on the formation of glue from fibers modified with hydroxylamine. The effect of target additives on hydroxylamine-modified fibers occurs mainly with amide groups. Complex formation between modified fibers and target additives also goes with nitrile groups. Therefore, most of the nitrile group in the spectrum of the glue is involved in complex formation (Fig. 7).

In destruction, cohesive breakdown of the adhesive and destruction of the adhesive bonds occur. Depending on the effect of this or that factor, thermal, thermoxidative and hydrolytic destructions are distinguished. It should be noted that the decrease in the strength of adhesive compounds occurs as a result of the influence of physical and chemical factors.

Water resistance of composite polymer glue. To determine the change in adhesion strength of the developed composite polymer adhesives, the water absorption of the glue samples was first studied and studies on the water resistance of the composite polymer adhesive were conducted.

Figure 8 shows the results of a study on the water absorption time of a modified composite polymer adhesive developed in water for 30 days.




1- unmodified, 2-starch, 3-dextrin, 4-wood flour

**Figure 8. Dependence of water absorption of polyacrylonitrile-based composite polymer glue modified with various additives on the duration of water absorption**

As can be seen from the curves 1, 2, 3, 4 in Figure 8, the water absorption of the samples increases in all cases over 30 days. Absorption of samples increases within 30 days. As a result of the subsequent re-

immersion of the samples, the water absorption is sharply reduced and remains practically the same when left in the water for 40 days, i.e. no increase in water absorption is observed. The water absorption of the unmodified sample is drastically different from that of the modified sample.

The lowest water absorption is observed in composite polymer glue modified with wood flour. The decrease in the water absorption index of the filled samples is explained by the appearance of three-dimensional sewn structures. This structure of the composition does not allow water molecules to enter the samples, which leads to a decrease in the water absorption of the material.

**CONCLUSION**

1. A scientific-methodological approach has been developed to create effective import-substituting composite polymer adhesive adhesives with high adhesion and cohesion properties based on organomineral ingredients in local and secondary raw materials and recommended for affixing excise brand labels of alcoholic beverages.

2. It was found that the improvement of physical-mechanical and adhesive properties of composite polymer adhesives depends on the nature, chemical composition and structure of the substances modifying the polymer adhesives.

3. The laws of influence of temperature on technological factors and quantity of various modifiers, and also on values of static exchange capacity (SAS) of polyacrylonitrile compositions are defined.

4. The addition of targeted modifiers to the adhesive composition was found to help improve the adhesion, strength and other physical and mechanical properties of composite polymer adhesives produced for use in the production of alcoholic beverages.

5. The optimal composition of KPK-1 and KPK-2 composite polymer adhesive adhesives with high adhesion and cohesion properties based on organomineral ingredients in local and secondary raw materials was developed and recommended for affixing excise brand labels of alcoholic beverages.

6. The obtained adhesive glue was found to meet the requirements for adhesives made using synthetic polymers and elastomers-based adhesives in the production of alcoholic beverages and other materials in all its properties.

7. A technology for obtaining adhesive composite polymer adhesives based on organomineral ingredients in local and secondary raw materials has been developed, which ensures the durability of excise labels on alcoholic beverages.

**References**

1. Tukhliyev G.A, Negmatova K.S, Babakhanova M.G, Soliyev R.Kh, Munavvarkhanov Z.T. Research of physical-chemical and strength properties of composite polymer adhesives based on local and secondary raw materials. // Journal of Critical Reviews, 2020. ISSN-2394-5125 No. 103. 326-329 (Scopus Impak Fact 2.1).

2. Negmatova K.S., Babakhanova M.G., Tukhliev G.A., Akhmedova D.U., Shodiev Kh.R. Development of scientific and methodological principles for the technology of obtaining composite polymer adhesives for labeling alcoholic beverages with excise stamps // Republican scientific and technical conference "Modern technologies for the production and processing of composite and nanocomposite materials", Tashkent, 2017, -P.182.

3. Babakhanova M.G., Tukhliev G.A., Adilova M.K., Ikramov N. Water-dispersive styrene-acrylic dispersions // New composite and nanocomposite materials: structure and application, Tashkent, 2018. -P. 144-146.

4. Babakhanova M.G., Tukhliev G.A., Negmatova M.I., Soliev R.Kh., Makhkamov D.I. Modified composite polymer adhesive // Resource-and energy-saving, environmentally friendly composite and nanocomposite materials, Tashkent, 2019, -S.205-207.

5. Babakhanova M.G., Tukhliev G.A., Nasriddinov A.Sh. Development of scientific and methodological principles of technology for obtaining composite polymer adhesives // National Academy of Sciences of Belarus. International Scientific and Technical Conference "Polymer Composites and Tribology" (Polikomtrib-2019) Gomel-Belarus, -S. 96.

6. Tukhliev G.A., Negmatova K.S., Abed N.S., Negmatov S.S., Rakhimov Kh.Yu., Babakhanova M.G. Investigation of the water resistance of a composite polymer adhesive based on polyacrylonitrile production waste as applied to stickers for labeling alcoholic beverages with excise stamps // Resource and energy saving, environmentally friendly composite and nanocomposite materials, Tashkent, 2019, pp. 288-290.

7. Negmatov S.S., Tukhliyev G.A., Babakhanova M.G. Research heating jf adhesive compounds // Procttdings of VIII international symposium on specialty polymers. Buketov University Karaganda. 2019.23-25 August, -S.33.

8. Tukhliev G.A., Babakhanova M.G., Negmatova K.S., Rakhimov Kh.Yu., Negmatov S.S. Water resistance of composite polymer glue filled with various target additives // International Uzbek-Belarusian Scientific and Technical Conference "Composite and metal-polymer materials for various industries and agriculture", Tashkent, 2020, pp. 113-114.

9. Babakhanova M.G., Negmatova K.S., Rakhimov Kh.Yu., Tukhliev G.A., Sultanov S.U. Modification of the langan composite polymer claying suvga chidamligini ўrganish // International scientific and technical conference "Improvement and implementation of innovative ideas in the field of chemistry and chemical technology." Fergana, October 23-24, 2020.-pp. 85-87.

10. Negmatov S.S., Babakhanova M.G., Tukhliev G.A., Negmatova K.S., Rakhimov Kh.Yu., Soliev R.Kh. Investigation of the influence of polyacrylonitrile on the modification of composite polymer glue // "Actual problems of innovative cooperation in improving the quality of higher education." International scientific online conference Navoiy, 2020, May 27, - P.15.

11. Tukhliev G.A., Babakhanova M.A., Negmatova K.S., Nasretdinov A.Sh., Rakhimov Kh.K. Development of scientific and methodological principles of the technology for obtaining composite polymer adhesives // VI Republican Scientific and Technical Conference of Young Scientists, dedicated to the memory of Corresponding Member of the National Academy of Sciences of Belarus S.S. Pesetsky "New functional materials, modern technologies and research methods-IMMS NAS of Belarus" Gomel, 2020. - P.70-72.

12. Babakhanova M.G., Tukhliev G.A., Negmatova K.S., Rakhimov Kh.Yu., Soliev R.Kh. Modification of urea-formaldehyde resin with polyarylonitrile fiber // Russian House of International Scientific and Technical Cooperation Russian Chemical Society. DI. Mendeleev "New polymer composite materials Mikitaevskie readings" Materials of the XVI International Scientific and Practical Conference-Nalchik, 2020. - P.53-56.