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CHEMICAL MEANS OF PROTECTING NONMETALLIC MATERIALS FROM ATTACK BY MICROORGANISMS

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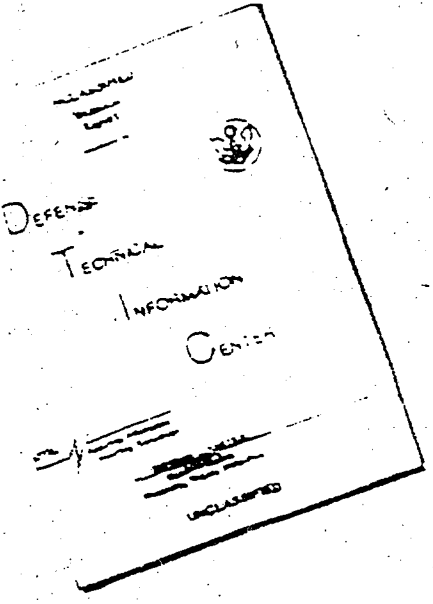
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CHEMICAL MEANS OF PROTECTING NONMETALLIC MATERIALS FROM ATTACK BY MICROORGANISMS

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CHEMICAL MEANS OF PROTECTING NONMETALLIC MATERIALS FROM ATTACK BY MICROORGANISMS

At the present time it is established that practically all nonmetallic materials can be attacked to one degree or another by various microorganisms. The attack of various materials occurs especially rapidly under tropical conditions at relatively high temperature and air moisture.

Materials of plant and animal origin, and also the products of their digestion, are most easily attacked by microorganisms. Various synthetic polymeric materials and even optical glass [1, 13] also undergo attack by microorganisms.

Unstable materials include: wood, paper and cardboard, nitrocellulose[1], cuprammonium rayon, biscose rayon[1], polyvinyl acetate[1], wool[2], leather[3], refined, unvulcanized, natural rubber[15], hevea latex[1], guayule latex[1], smoked sheets[6], gutta-percha[6], regenerated rubber[1], neoprene with nutritive additive[6], butadiene-styrole SK[1], Buna-S[1], Buna-H[8,12], Isobutylene-Isoprene SK[6], Unvulcanized[8] and vulcanized[16] Thiokol, Phenolo-anilino-formaldehyde resin[1], alkyd resin[12].

The following materials are stable: mixed neoprene rubber[6,12,15], silicone resin[1], orlon [17], polymethacrylate[12], phenolo-formaldehyde resin [12], urea-formaldehyde resin[12,18], teflon[19], polystyrene[12,19], polyisobutylene [12,20]. According to one report neoprene-polyvinylchloride is a material which is resistant to the effect of microorganisms, that according to others[8,10] - it is not resistant.

In many cases the action of microorganisms does not have a substantial effect in changing the durability of the material, but has a negative effect on its external appearance, and also on its electrical properties[1,14]. Thus, the specific volume electrical resistivity of certain insulating materials (polyvinylchloride, plasticized resin and perchlorvinyl coatings of various resins, lacquer coatings and many other materials) drop 100-fold under the influence of molds.

It should be noted that the resistance of plastics to microorganisms has still not been studied adequately in recent times and there is much data indicating the vulnerability of various materials which were previously considered to be completely resistant. It has been established positively that all articles from very different polymers which contain cellulose as filler and other organic substances capable of serving as nutritive media for molds and other microorganisms are very vulnerable to microorganisms[1].

The prevention of growth of microorganisms on various articles can be accomplished by many methods, of which the following have practical importance: application of materials resistant to the action of microorganisms, and addition of special antiseptics which kill microorganisms or inhibit their growth to materials.

It is necessary, however, to indicate that materials which possess absolute resistance have still not been found at the present time and frequently growth of molds is observed on relatively resistant materials which have no nutritive medium at all for fungi, and are contaminated by organic substances from the atmosphere and soil[1].

Due to the most extensive distribution mentioned above, protection of various materials from attack by microorganisms was obtained by means of addition of special antiseptics to them.

The most extensive use of antiseptics was attained for protection of wood- material which is used in extremely different fields of industry and construction. Without touching on questions of protection of wood, which were considered in print several years ago[21], that the rate of manufacture of antiseptics for wood has continually enlarged. This is quite convincingly clear in the case of Pentachlorphenol, whose use for antiseptic treatment of wood in the USA increased from 126.7 tons in 1945 to 5,138.1 tons in 1958[22].

We note that antiseptics which are effective for one type of microorganism may appear to have little effect against others. The materials into which the antiseptics are introduced, and also the conditions of introduction of the antiseptics and the treatment of the material appear to have a great influence on the effectiveness of the antiseptics. There are indications in the literature that even the solvent may have an effect on the activity of the antiseptic[23], which is especially important when using antiseptic for paint and varnish coatings. The pH of the medium also has a substantial effect on the activity of antiseptics[24].

Usually the following general requirements apply to antiseptics: the presence of fungicidal and bactericidal effect against a wide circle of microorganisms, chemical and physical stability, absence of odor and color, low volatility and low solubility in water, etc. Moreover, an antiseptic introduced into a given material, should not effect its chemical and physical properties, should not harm the quality of the article, or change the technology of its preparation and harm the working condition. Antiseptics should be sufficiently inexpensive and have comparatively low toxicity for humans and animals. Antiseptics which simultaneously satisfy all the enumerated requirements have not yet been found, although more than 3,000 compounds have been published in the literature, which have been tested at various times in very different materials. Practical application has been obtained for an extremely limited number of substances.

In a short review it is not possible to give even a simple list of the antiseptics which have been studied up to the present time, and we are attempting to mention only certain preparations which have sufficiently great practical application. At the present time a very great number of classes of organic compounds have been studied as antiseptics for various nonmetallic materials, including phenols, nitrocompounds, halogen derivatives of hydrocarbons, simple and complex ethers, salts of various organic acids, heterocyclic compounds, quinones, amides of acids, organic compounds of mercury and tin and many others[25a].

Of the phenols, the most widely used as antiseptics for nonmetallic materials are the tri-tetra- and pentachlorphenols, which are used for protecting paper and cardboard[1,26-28], cotton and other fibers of plant origin[29-35], plastic substances[15,37], paint and varnish coverings[36,38-43], leather[44,45], certain other materials[1] from attack by microorganisms. Usually these phenols are introduced in quantities from 0.4 to 2% of the weight of the dry material being preserved. In several cases in order to increase resistance in the presence of moisture antiseptics are introduced along with hydrophobic additives (most frequently bitumen, paraffin or synthetic resins).

Of the substances of the phenol type 2,2'-dioxy-5,5'-dichlordiphenylmethane[1,46-48], n-nitrophenol[1,49,50], oxydiphenyl[50,56,57], β-naphthol[49], have also received some application as to antiseptics. Thus, n-nitrophenol proved to be a good antiseptic for leather and natural and synthetic rubbers.

It is appropriate to note that as a result of prolonged action of sunlight almost all halogenophenols impregnating plant fibers, break down with the formation of hydrogen chloride, which results in some reduction of the durability of the fiber[1].

Along with free phenols, their salts with various metals are also used: copper, zinc, silver, cadmium, etc. Interesting results[1] were obtained, in particular, with silver pentachlorphenolate, which was used for protection of cotton fabric in Florida (USA) over a period of two years in air and for more than three weeks in soil. After remaining for two years in open air the fabric treated with 1% silver pentachlorphenolate, lost only 11-13% of its original durability, while control samples had centergraded by 90-100%. Somewhat poorer results were obtained by treatment of fabric with copper pentachlorphenolate, inasmuch as under the same conditions after two years the durability of the fabric was reduced by 25%, and fabric treated with free pentachlorphenol, had deteriorated by 77-95%.

Mixtures of various phenols[1], and also mixtures of phenols with ketones and halogen derivatives of aromatic hydrocarbons[1,59] possess increased activity. 4,4'- dinitrodiphenylcarbonate[60] has also been proposed as active antiseptic for leather.

Copper 8-oxyquinolate, which is used for protection of textile products[2, 56, 61-69] including fire hoses, cotton and parachute fabrics and various other materials used in airplane construction[23,56], lacquers and paints[70-72], and plastic substances[57,73], have received wide application for antiseptic treatment. The addition of copper 8-oxyquinolate, and also other copper compounds to natural rubber leads to rapid aging, while certain synthetic rubbers in this case proved to be resistant[57].

Copper 8-oxyquinolate is added in a quantity from 0.5-8-10% depending on the material. It may be added by various methods, for example, precipitation of the material as a result of reaction with inorganic copper compounds with salts of 8-oxyquinolate, or in the form of solutions in various organic solvents[69] or in the form of its aqueous suspension, and also an emulsion of its solutions[1]. The presence of a color is a substantial drawback of this antiseptic (as it is with almost all other organic copper compounds), which to a certain extent limits the possibility of its use.

Recently, the use of 8-oxyquinolates of cadmium[74], lead[75], and other metals as antiseptics for various materials has been patented.

The use of copper naphthenate, use production in the USA in 1958 exceeded 1,000 tons[76] has reached significant levels. Since copper naphthenate has an unpleasant odor and stains materials, it is used mainly for treatment of cruder articles[1]. Along with copper naphthenate, the use of naphthenates of other metals and their mixtures with copper naphthenate has been proposed[1]. There are data in the patent literature on the high activity of copper salts of various other organic acids as well (for example, furancarbonate and furandithiocarbamate[77,78], and also stearic, palmitic, and oleic acids[1]).

Dinitrofluorobenzene, dinitroflurobromobenzene, 1-fluoro-3-chloro-4,6-dinitrobenzene and 1,3-difluoro-4,6-dinitrobenzene[24,37] are interesting antiseptics for various aviation materials and especially plastics. These compounds are quite soluble in organic solvents and in many high polymers, are colorless and have practically no odor. They significantly surpass phenols and oxyquinolates in microbiological activity. Fluoronitrobenzene is also suggested for use in protecting leather[24,57]. One must note, however, that it is rather complicated to obtain these compounds (with the exception of 2,4-dinitrofluorobenzene), as a result of which their cost is higher than halogenophenols and 8-oxyquinolate derivatives.

Several other halogenonitrobenzenes have proved to be active fungicides for the protection of leather[79].

Organic compounds of tin, which are close to organic compounds of mercury in the strength of their effect are new, promising antiseptics for nonmetallic materials [66,80-86]. The minimal concentrations of several organic compounds of tin, which completely inhibit the growth of fungi required for characteristic fungicidal activity of this group of substances are presented in the table[82,83,92].

Fungicidal Activity of Several Tin Compounds

Compounds	Minimum concentration for full stoppage of growth			
	Botrytis allii	Penicillium itallicum	Aspergillis niger	Rhizopus nigricans
Trimethyltinacetate	[illegible]	20	[illegible]	[illegible]
Triethyltinacetate	1	2	5	2
Tripropyltinacetate	0.1	[illegible]	1	1
Triisopropyltinacetate	[illegible]	[illegible]	[illegible]	[illegible]
Tributyltinacetate	0.1	[illegible]	0.5	0.5
Trihexyltinacetate	1	10	2	[illegible]
Trioctyltinacetate	[illegible]	100	10	10
Triphenyltinacetate	2	1	0.5	1.0
Diethylphenyltinacetate	1	2	5	2
Diethyl-n-Bromophenyltinacetate	0.2	0.5	1	1
Diethylbutyltinacetate	0.1	1	0.5	0.5
Diethylhexyltinacetate	0.06	0.5	0.1	0.1
Diethyloctyltinacetate	0.02	0.5	0.1	5
Ethyldipentyltinacetate	0.2	1	[illegible]	0.5
Phenylmercuriacetate	0.5	0.5	0.5	5

Organic tin compounds are good antiseptics for wood[82,83], cotton and jute[84], paints[85], paint and lacquer coatings[80], paper[81] and other materials. Organic tin compounds began to be used as agents for protecting plants from diseases[93,94] and for preventing slime formation in the paper industry[86,88], as effective substitutes for organic mercury compounds, less toxic for humans and domestic animals[95].

In industry the acetate and oxide of tributyltin has had the widest application, and in agriculture - triphenyltin-acetate.

It is interesting to note that several organic compounds of tin are stabilizers for various plastic materials, increase their thermal stability, prevent yellowing and increase the duration of service in sunlight.[87-90]. The stabilization of polyvinylchloride resin by organic tin compounds is of particular interest.

Organic compounds of mercury[1,20,36,66,96-103], which are recommended for treatment of textile materials[1], wood and paper[1,20,66,96, 97], plastic materials[1,20,98], resin[99], lacquers and paints[100,102], are used as universal antiseptics for many types of nonmetallic materials.

These compounds include phenylmercuriacetate, -phthalate, -sulfamide, -oleate, pyridylmercurichloride, alkylmercurioleate, ethylmercuriphosphate and several others. Usually organic compounds of mercury are used in quantities from 0.1 to 1% of the weight of the treated material, considering dry matter[1]. The general disadvantage of these compounds of mercury is their high toxicity for persons and animals, which is aggrevated by their ability to accumulate in the organism and to cause serious chronic poisoning. The high toxicity of organic mercury compounds for humans leads to the necessity of taking special precautionary measures both in their production and in their use, which significantly increases the cost of production.

A negative property of organic mercury compounds is also their low stability in relation to soil microorganisms, by which they are broken down according to the following scheme: $2RHgX + 2H \quad R_2Hg + Hg + 2HX$

Inorganic mercury compounds[102] have also been proposed as antiseptic for nonmetallic materials, but their effect is significantly weaker as a result of which they must be introduced into the treated material at higher quantities than organic compounds.

Salicylanilide is an intersting universal antiseptic with low toxicity for humans, which is recommended for protection of paper[126], textile materials[1]; plastics[20,106], lacquer and paint coatings[104,105], resins and other materials[1].

Salicylanilide is poorly soluble in water, rather stable to heating and comparatively inexpensive. However, in order to obtain a good protective effect, salicylanilide must be introduced into the treated material at high quantities (3-8%) which in many cases shows up negatively in the properties of the treated material. An especially pronounced negative effect of salicylanilide appears in lacquer coatings in which the preparation sometimes separates in the form of crystals.

There is some use of quaternary ammonium salts[107-111], which possess comparatively weak fungicidal activity, but more bactericidal. In view of its good solubility in water its application is expedient only in cases when the treated material is not subjected to systematic effect of water.

Along with the substances mentioned above as antiseptics for nonmetallic materials, 2,3-dichloronaphthoquinone[1,31], rhodanine[112], salicylaldoxime[113], certain pyridine derivitives [114,115], zinc thiocyanate[116], n-toluolsulfamide[1], amides of various other acids[117], and many other compounds[1,25a,36,118,119] are also suggested.

The use of antiseptics for protection from microorganisms has great significance not only for protection of various nonmetallic materials, but also for other purposes in certain fields of industry. Protection from biological growth is especially important in the paper industry. According to data of various investigators biological overgrowth of apparatus in paper factories in the USA results in annual losses of about 25 million dollars[1]. Organic mercury compounds, their mixtures with phenols and oxyquinoline[120-122], and also the hydrate of tributyltinoxide[86,88], have been used recently as agents for combating biological overgrowth in the paper industry.

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13. ABSTRACT

Various antiseptics are suggested for protection of susceptible materials of plant and animal origin as well as certain synthetic polymers from attack by microorganisms. Antiseptics should possess fungicidal and bactericidal properties against a wide range of microorganisms, chemical and physical stability, freedom from odor and color, low volatility and low solubility in water, and should not effect the chemical or physical properties of the treated material or the quality of the article. In addition they should be inexpensive and have low toxicity for humans. Antiseptics discussed include: phenols, 8-oxyquinoline derivitives, naphthenates, nitro- and halogen derivitives of benzene, organic and inorganic mercury and tin compounds, and quaternary ammonium salts.

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