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PROMISING PREPARATIONS FOR ACARICIDE-REPELLENT FINISHING OF TEXTILE MATERIALS

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Abstract. *The article considers the current state of the market for acaricidal repellents and the methods of human protection by these compounds. The authors give the description of acaricidal repellents of natural and synthetic origin. The work proposes the domestic pyrethroid as method for human protection against insects by encapsulating it in microcapsules and then applying to textile material. Also the work reviews domestic and foreign technologies for treating textile materials with acaricidal repellents.*

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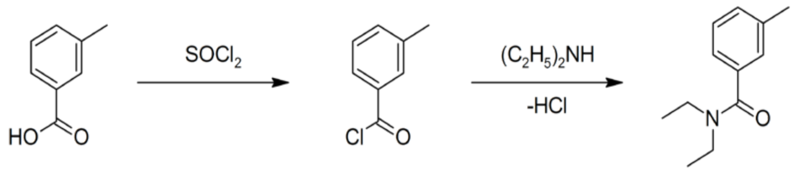
Introduction

An important task of modern textile production is to produce high-tech technical fabrics with innovative styles of finishes. These finishes are virulicide, antibacterial, acaricidal repellent and masking finishes with infra-red effect.

Clothing with an acaricidal repellent finish is important for professions such as geologists, oil workers and military personnel operating in forest areas. Protecting humans from mosquitoes and ixodid ticks (*Ixodidae*), carrying the tick-borne encephalitis virus, is urgent during the spring and summer. There are several ways to solve this problem. The first is to vaccinate the population, which can be complicated by allergic reactions; the second is to pollinate forests and parks with acaricides, which are quite toxic to the environment; the third way is to use special suits for mechanical and chemical protection of humans against insects. Both natural and synthetic repellents can be used for acaricide-repellent finishing of textiles.

Synthetic chemicals applying on human skin, clothing or other surfaces are called synthetic repellents. They prevent insects from entering the treated surface. The potential for contact with human skin is reduced by synthetic repellents feature to make the surface of the clothing unpleasant and unattractive to insects. Repellents also help prevent and control outbreaks of diseases transmitted by insects: malaria, *amarillic typhus amarillosis*, *Japanese encephalitis, Dengue Haemorrhagic Fever* (DHF), etc. The structure of repellents contains active groups, such as the cyanogroup, which can repel insects by blocking their olfactory senses, which are responsible for detecting carbon dioxide and lactic acid produced by sweating [1].

One of the best known and most commonly used repellents is diethyltoluamide (DETA), which is a light oil-like liquid with a slight aromatic odour. It was developed for the US Army in 1946 to protect soldiers in regions with a high insect presence. In the USA it was registered for civilian use in 1957. Since 1965 it is on the market as part of repellents for individual use. The repellent effect when applied to the skin against mosquitoes lasts 10-12 hours and against blackfly up to 6 hours. When sprayed on clothing, the protective effect against mosquitoes can last up to 2-3 days, while application of 20-35% emulsion provides protection for 30 days. The synthesis of DETA follows a pattern:



Methatoluic acid is converted to chloranhydride by thionyl chloride/ After the chloranhydride treats with diethylamine produce the diethylamide of m-toluic acid – diethyltoluamide (Fig. 1) [2].

The toxicity of DETA to the environment and living organisms is limited, but it is known about its effects on the brain. Little information is available on the toxicity of DETA to aquatic invertebrates. The substance is unique in its ability to mask the sensory perception of lactic acid on the skin, making it unattractive to insects such as mosquitoes and ticks. The disadvantages of this compound are sharp odour, subchronic toxicity, mutagenicity, reproductive and neurological toxicity [3, 4].

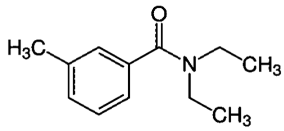


Fig. 1. Structural formula of DETA

N,N-diethylphenylacetamide (DEPA) is repellent for widespread use, too (Fig. 2). According to toxicological studies, DEPA does not cause skin irritation or photochemical reactions after short-term exposure to UV light, being a safer alternative to DETA [5].

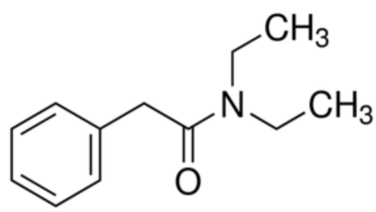


Fig. 2. Structural formula of N,N-diethylphenylacetamide

One well-known synthetic insect repellent is picaridin (Fig. 3). Its protection time is identical to that of DETA at similar concentrations. Picaridin is recommended to be applied to human skin and/or clothing. It protects against mosquitoes, flies, ticks and fleas. The advantage of picaridin is absence of damage to plastics and synthetic fabrics.

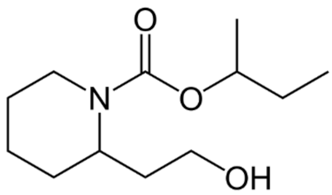


Fig. 3. Structural formula of picaridin

The exact mechanism of action of picaridin is unknown. No dermal, organ-specific or reproductive toxicity at doses up to 200 mg/kg body weight was detected in animal studies, nor were any teratological, developmental or neoplastic abnormalities detected. The concentration of this substance determines the time of protection against insects. The concentrations of picaridin 7, 5, 10 and 15% are at the market. When its concentration is increased from 7.5 to 15%, the protective time varies from 2 to 4 hours [6]. The disadvantage of this substance is its short protection time against insects.

Metoflutrin ("SumiVan") (2,3,5,6-tetrafluoro-4-(methoxymethyl)benzyl 2,2-dimethyl-3-(prop-1-en-1-yl)cyclopropane carboxylate (Fig. 4) can also be used as a repellent.

This substance is a volatile insecticide produced by Sumitomo Chemical LTD (Japan). It characterized by high paralysing activity for mosquitoes. It is available in forms of spirals, liquid fumigators, pepelators and dispensers [7, 8].

Methoflutrin has high repellent properties against the *Aedesalbopictus*and the *Aedestaeniorhynchus*[9].

The disadvantage is high aqueous toxicity.

The major metabolites of methoflutrin were investigated using three different aquatic species - fish and algae: *Pimephalespromelas*, *Daphnia magna* and *Pseudokirchneriellasubcapitata.* The high toxicity has been detected for all metabolites [10], suggesting the substance is unsafe for the environment and humans.

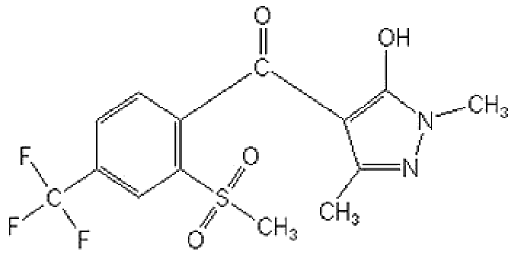


Fig. 4. Structural formula of metoflutrin

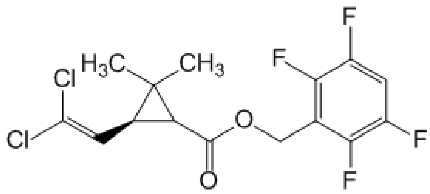


Fig. 5. Structural formula of transfluthrin

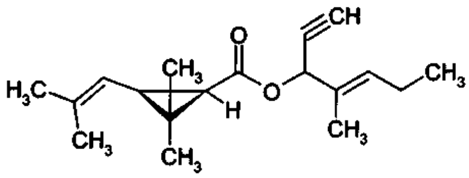


Fig. 6 Structural formula of D-Empentrine

The substance having the acute action against blood-sucking insectss is Transfluthrin, 2,3,5,6-tetrafluorobenzyl (1R,3S)-2,2-dimethyl-3-(2,2-dichlorovinyl)-cyclopropane carboxylate (Fig. 5) (Bayer, Germany). It causes paralysis of the insect in case of contact and fumigation. Also it has rapid action during contact and inhalation activity against mosquitoes, flies, cockroaches and whiteflies. It is an effector of presynaptic voltaic-dependent sodium channels of nerve membranes, by mechanism of action, causing insects' knockdown effects.

Transfluthrin is used both alone and together with different pyrethroids in electrofumigators and products in aerosol packages to control flying insects. This substance, due to its increased volatility, can be used in antimoth plates [11, 12].

D-Empentrine ("Vaportrine") (Fig. 6) is a pesticide, an insecticide used to control flies and mosquitoes indoors. The Vaportrin is produced in the form of plates designed to protect fur, wool and wares from damage by keratophagous insects.

The substance is highly active, exceeding the effectiveness of peritroids such as, for example, allethrin by a factor of 2.5. The mechanism of this drug action is based on disruption of sodium ion metabolism, leading to the release of large quantities of acetylcholine as the nerve impulse passes through the synaptic cleft. In the Russian Federation such substances are on the market since 2004 in the form of plates and liquids forelectrofumigators [13]. The disadvantage of this substance is the occurrence of allergic reactions in humans and animals.

The repellents described are used quite effectively abroad and in our country. However, the disadvantages of these substances, such as their short duration of action, high volatility, ability to cause adverse and allergic reactions in humans and animals, made scientists to look for ways of replacement. In this respect, synthetic pyrethroids, which are analogues of natural fast evaporating pyrethrins, are significant.

Pyrethroids are synthetic insecticides analogous to natural ones, in particulary*Pirethrumcinerariifolium* or *Tanacetumcinerariifolium* from Kenya, Rwanda, Ecuador and Tanzania [14]. Pyrethroidsare optically active high boiling liquids, soluble in the most organic solvents, practically insoluble in water; easily oxidised in air and light. They are esters in their chemical nature.

First-generation pyrethroids include chrysanthemic acid esters (Fig. 7) [15].

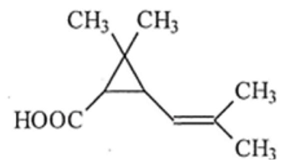


Fig. 7. Chrysanthemic acid formula

These pyrethroids have high insecticidal activity. The disadvantages are their easily oxidation on the natural light [16]. These substances are used in mosquito repellents and plates (Raptor, Fumitox, etc.) and in the form of sprays. The use of such pyrethroids by spraying may cause allergic reactions.

The second generation of pyrethroids developed in the 1960s and 1970s. They were already more stable to photo-oxidation (3-(2,2-dihalogenvinyl)-2,2-dimethylcyclopropane-carboxylic acid esters - permethrin, alpha-cypermethrin, deltamethrin (decamethrin, "decis") and fenvalerate, a pyrethroid that contains no cyclopropane ring [17]. These pyrethroids have a wide spectrum of action and are effective at very low consumption rates. These substances have a more powerful insecticidal effect and are used for the control of domestic insects and for the treatment of fabrics and packaging materials.

Third-generation pyrethroids are cyhalothrin, flucitrinate, fluvalinate, tralmethrin, cyfluthrin, fenpropatrin, bifetrin, cycloprotrin, and etophenprox. The most common is cigalothrin (it is 2.5 times more active than deltamethrin). They are known to be highly effective against mites, while at the same time being toxic to bees, birds and fish [18, 19].

Nowadays, a second-generation pyrethroid - permethrin (antiparasitic agent: insecticidal, acaricidal, antipediculosis) is used as mosquito and tick repellents and for fabric repellent technology (Fig. 8) [20]. It has high protective efficacy against mosquitoes and flies [21].

The insects' knockdown effect requires direct contact of the substance and the insect. As a result there is a blockage of sodium channels, inhibition of acetylcholinesterase activity and paralysis of the insect. There were no teratogenic effects in animal studies [22].

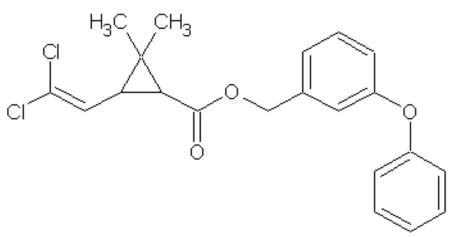


Fig. 8. Structural formula of permethrin

Permethrin is actively used for repellent finishing of textiles abroad due to its multiple effects on insects (repellent, disorienting and lethal) [23]. The disadvantage is it can cause hyperactivity, aggressive behaviour and tremors of animals (24). There is a risk of an insect stinging a person before it dies. Fabric finishing technologies and permethrin-based products are expensive and imported

The most effective of the permethrins is alpha-cypermethrin [mixture of cypermethrin isomers (1:1): (S)-α-cyano-3-phenoxybenzyl ether of (1R)-cis-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropancarboxylic acid and (R)-α-cyano-3-phenoxybenzyl ether of (1S)-cis-3-(2,2-

dichlorovinyl)-2,2-dimethylcyclopropancarboxylic acid], which is used in agriculture and in private households medical and sanitary practice for disinsection.

Alpha-cypermethrin (ACP) is a white crystalline powder with a melting point of 78-81 °C, with density d = 1.86 g/cm^3, a low-volatile substance (Fig. 9).

Alpha-cypermethrin (technical) was registered as an insecticidal substance for the production of insecticidal agents in 2002 (Protocol No 135 of 19-20 July 2002). It possesses high acaricidal activity. It is 28 times more active than permethrin, 2-5 times faster than cypermethrin, and causes ticks' knockdown effect [25].

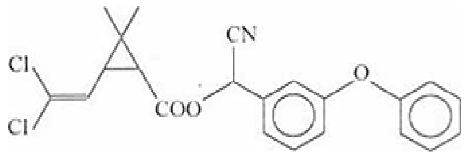


Fig. 9. Alpha-cypermethrin formula

The use of lotions, spirals, sprays containing chemicals such as DETA, permethrin, cypermethrin and alpha-cypermethrin leads to unavoidable contact of volatile organic substances and repellents with human respiratory tract, causing allergic reactions.

This problem can be solved by using textile materials treated with natural and synthetic repellents.

In order to obtain a stable acaricidal-repellent finish with prolonged release of alpha-cypermethrin it is necessary to use it in the form of microcapsule. The microencapsulation method allows keep the stability in contact with other substances, as well as light fastness and absence of side effects on the body [26-30].

Protective clothing should be comfortable and safe, with certificates of conformity and health and safety approvals [31, 32].

In order to achieve the maximum effect of human protection against blood-sucking insects, a specific model of anti-encephalitis clothing with a high degree of acaricidal protection by giving it an acaricidal-repellent finish is necessary.

Existing technologies for treatment of fabrics include spraying the preparation onto the fabric surface, the use of two-step technology developed by G.A. Krestov Institute of Solution Chemistry of the Russian Academy of Sciences (ISC RAS) using diethyltoluamide as a repellent [33] and technology based on Sanitized AM 23-24 (by Clariant, Switzerland) [34]. Permethrin is also used in textiles at different stages of production: during fibre preparation, yarn preparation or at the final product stage [35].

These technologies are successful, but have disadvantages: the possible ingress of the substance into the human respiratory tract during spraying, the use of drugs that can cause side effects in humans (DETA), the need for complex sealed hardware design and volatile organic substances as solvents for repellents, the use of an expensive imported product based on permethrin (Sanitized AM 23-24).

We consider the process of combining the treatment of textile material with permethrin and MCT-β-CD cyclodextrins. The method is ecofriendly. Cotton fabric is treated separately with cyclodextrin and then a permethrin-based insect repellent. The treated fabric is stored in a hermetically sealed container [35]. This method of processing is effective, but costly and complex in terms of hardware.

In some cases, a method of applying the mosquito repellent N-N-diethylbenzamide to textile material by textile printing is used [36]. This method is promising, but there is insufficient evidence to support its effectiveness in production. Also we should take into account the use of thermosetting resin precondensate for fixation of this substance on the textile material. The challenge of creating a sustainable acaricidal repellent finish is both in the rational choice of the least toxic to humans and the environment product, which terminates insects and fixes effective on the textile. The repellent release from the surface of the textile material must be prolonged over time and the effect of the finish must be washable.

Alpha-cypermethrin is suggested as the most effective acaricide with high insecticidal properties.Also it is relatively harmless for humans and animals and inexpensive. The encapsulation of alpha-cypermethrin provides stability in possible contact with other chemicals, light fastness and safety for humans [37].

Department of Chemical Technology of Fibre Materials of Ivanovo State University of Chemistry and Technology (Ivanovo, Russia) in cooperation with the company SPECIAL TEXTIL Association (Shuya, Russia) developed a technology for the antaricidal and repellent treatment of textile materials with a preparation based on alpha-cypermethrin. The product is a dispersion containing microcapsules composed of a core with alpha-cypermethrin dissolved in a non-toxic oil solvent (oxyethylated rapeseed oil) and a shell formed from oppositely charged cationic and anionic polyelectrolytes and surfactants. The textile material is treated with the obtained microcapsule dispersion, followed by convection drying and layer-by-layer immobilisation of the capsules on the textile material using differentially charged cationic and anionic polyelectrolytes or the fixative "Texoklen BZU-M", followed by contact drying.

The fabric samples are used to make "Barrier-Insecto" protective clothing with additional mechanical protection in the form of tick traps. The technology developed was successfully tested by production. The clothing provides high protection against ticks (98.2%, with a regulatory value of at least 98%), and 97.5% against blackfly with a regulatory value of at least 95%) [32, 38-40].

Conclusions

The paper reviewed a wide range of acaricidal repellents and methods of protecting humans from insects. It also analysed the methods of imparting acaricidal-repellent properties to textile materials and considered the advantages and disadvantages of existing fabric finishing technologies. The authore proposed the technology of acaricide-repellent finishing of textile materials based on microencapsulated alpha-cypermethrin. This technology is successfully implemented in the production of the "SPECIAL TEXTILE Association" (Shuya, Russia).

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