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Review

WATERLESS CLEANING COMPOSITIONS WITH DISINFECTION PROPERTIES: EFFICACY AND ENVIRONMENTAL ASPECTS

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The microbial ecology of different indoor environments determines the human microbiome. Hence, cleaning and disinfection of indoor environments like hospitals, apartments, automobiles, etc. are of great importance. Nonaqueous surface cleaning preparations (SCPs) are often used for this purpose. A cleaning composition may contain the following compounds: primary surfactant, cosurfactant, solvent, cosolvent, organotropic (organic solubilizing) agent, hydrotropic (water solubilising) agent, water and salts, and special additives. In this paper, the efficacy of complex preparations is discussed, focusing on the chemical composition and testing methods. Particular attention is paid to quaternary ammonium compounds, i.e. surfactants with disinfection properties, which are known to act as antistatic agents, detergents, oil-in-water emulsifiers, corrosion inhibitors, and lubricants. Specificity of target microorganisms, physicochemical properties of surrounding media and treatment mode are the main factors affecting the efficacy of disinfection. Due to the enormous economic importance and massive worldwide use of surfactants and disinfectants, their environmental impact needs to be evaluated and controlled. Increased knowledge and better understanding of the antimicrobial capacity of disinfectants are essential to optimise sanitation procedures, to reduce costs, environment waste and to improve shelf life.

Key words: *biodegradability, quaternary ammonium compounds, minimum inhibitory concentration, surface cleaning, toxicity.*

AREAS FOR APPLICATION OF SURFACE CLEANING PREPARATIONS

Cleaning and disinfecting the indoor environments represents a huge area of current research. The studies are focused not only on buildings of the health care sector. The microbial ecology of different indoor environments determines the human microbiome. Therefore, potential strategies for controlling the presence of human pathogens in apartments, automobiles and other built environments are of a great importance worldwide (Klepeis *et al*., 2001; Kembel *et al*., 2012; Stephenson *et al*., 2014).

Sources of microbes for indoor microbiomes are mainly from outside air or the human skin (Pakarinen *et al*., 2008; Rintala *et al*., 2008; Grice and Segre, 2011). In particular, staphylococci frequently colonize human skin and mucosal surfaces, and thus are transmitted to surfaces that humans come into contact with (Stephenson *et al*., 2014; Foster, 2009; Payne *et al*., 2013).

Stephenson and coworkers (2014) studied the most highly colonised locations of pathogens in automobiles. These areas were suspected to have frequent touching by the occupants, such as locations on the steering wheel, the gear shifter, door handles and window switches, and the centre console near the beverage holder (Stephenson *et al*., 2014).

For hard surfaces, washing with soap and water and rinsing can significantly reduce bacterial loadings (Cogan *et al*., 2002). Disinfection in hospitals is recommended only for surfaces in frequent contact with hands and skin of patients and personnel, as repeated disinfection of other areas is unnecessary and leads to allergic symptoms in health care workers (Dascher *et al*., 2004; Henry, 2011). Disinfection of wet cleaning cloths is necessary, because the probability of contamination and transfer is high (Bloomfield and Scott, 1997).

CHARACTERISTICS OF TARGET MICROORGANISMS

Microorganisms vary considerably in their response to disinfectants. Bacterial spores are the least susceptible, followed by mycobacteria and then by Gram-negative bacteria, notably pseudomonads. Gram-positive cocci, including antibiotic-resistant staphylococci are readily inactivated by disinfectants. Enterococci are also susceptible but somewhat less so than staphylococci (Maillard, 2002; Russel, 2002).

It is desirable to test a broad spectrum of organisms for their susceptibility to disinfectants. In this respect, additional attention should be paid to: i) the high levels of antibiotic resistance in plant-colonising enterococci; ii) the fact that *Burkholderia cepacia* can cause both plant and human infections; iii) the ability of protozoa to act as reservoirs of animal and human pathogens (Dixon, 2002).

Hospital isolates are often more resistant to biocides than laboratory or 'standard' strains. Overusage of antibiotics (e.g., animal feedstuffs) and disinfectants (e.g., domiciliary environments) can lead to selection for antibiotic-resistant bacteria, a potentially serious situation (Russel, 2002). As reported earlier, both antimicrobials, i.e. biocides and antibiotics, might develop shared resistance mechanisms, e.g. efflux pumps, permeability changes and biofilms (Anonymous, 2009). Resistance to these compounds may be acquired either by mutation or by the acquisition of genetic elements (plasmids, transposons).

There are two major mechanisms whereby bacteria show resistance to disinfectants. The first one is based on impermeability of the outer bacterial cell layers and is formed in bacterial spores, mycobacteria and Gram-negative organisms. Alternatively, some organisms, such as *P. aeruginosa*, can efflux some agents such as triclosan, chlorhexidine and quaternary ammonium compounds (QACs) (Chuanchen *et al.*, 2001; Poole, 2001; Russel, 2002).

By analogy with antibiotics, disinfectant rotation policies are practised in some hospitals. Such policies attract strong opinions in support and against (Murtough *et al.*, 2002; Russel, 2002).

METHODS FOR EVALUATION OF DISINFECTANTS

Unlike sterilisation, disinfection is not sporicidal. A few disinfectants kill spores with prolonged exposure times (3–12 hours); these are called chemical sterilants. High-, intermediate- and low-level disinfectants differ in their antimicrobial spectrum and rapidity of action (Rutala and Weber, 2008).

Studies on efficiency of surface disinfectant cleaners carried out with test-microorganisms in suspension and not under practical conditions, can be considered as a strong limitation (Reichel *et al.*, 2014). In case the efficiency of a disinfectant applied to surfaces is based on counting the microbial survivors sampled in a liquid, then total cell removal from surfaces is seldom achieved. As a result, the efficiency of surface disinfection procedures can be overestimated (Grand *et al.*, 2011). Use of fluorescent dyes in microscopy can improve the assessment of cell viability, even for surface-associated cells (Davinson *et al.*, 2010; Bridier *et al.*, 2011; Grand *et al.*, 2011).

Some studies aimed at developing disinfection procedures at food processing plants have led to modification of the standard methods in order to adapt them to technological conditions. Thus, for a fish processing plant, bactericidal efficiency of common disinfectants against adherent cells on stainless steel surface was carried out with mixed culture of *Pseudomonas putida, Serratia liquefaciens* and *Shewanella putrefaciens* isolated from shrimp and fish processing plants (Duong, 2005).

Standard methods for determining bactericidal, sporicidal, fungicidal and yeasticidal effect of the chemical disinfectants and antiseptics on the surface and in suspension, are provided in EU documents (Anonymous, 1997a; 1997b; 2001).

FACTORS AFFECTING THE EFFICACY OF DISINFECTION

Among the factors influencing an efficacy of disinfection, specificity of target microorganisms, physicochemical properties of surrounding media and treatment mode are considered as most important.

Target microorganisms. Susceptibility of microorganisms to the disinfectant is dependent on the type, source, concentration of microorganisms, as well as the presence of biofilm. Activity of a disinfectant might also differ between different strains of the same species (Maillard, 2002).

Media. The pH level, hardness, salinity, the presence of organic or inorganic interfering material, physical nature of the object (e.g., crevices, hinges, and lumens) can sufficiently influence the outcome of disinfection procedure. The presence of divalent cations increases susceptibility to QAC (Crismaru *et al.*, 2011). Quaternary-containing disinfectants are affected by water hardness and less affected by organic matter (Anonymous, 2000; Duong, 2005).

Treatment mode. Sufficient contact time is critical to ensure disinfection. For example, the standard suspension methods suggest a contact period with disinfectant of 5 min and 15 min for tests with bacteria and fungi/yeast cultures, respectively. An appropriate neutraliser should be used before counting the number of cells surviving the test disinfection procedure (Anonymous, 1997a; 1997b).

CHEMISTRY OF SURFACE CLEANING PREPARATIONS

Waterless cleaning preparations. A formulated waterless cleaning composition may contain the following compounds: primary surfactant, cosurfactant, solvents, cosolvent, organotropic (organic solubilising) agent, hydrotropic (water solubilizing) agent, water and salts, and special additives. The water content of most cleaning aids ranges from 5% to 15%. Additives, such as softening, retexturing (sizing), antistatic, oxidising (bleaching), disinfecting, and optical brightening agents, can be beneficial in cleaning aids.

The most important and usually the most effective constituent of a cleaning aid is a surface-active substance. Historically, six classes of surfactants have been used in waterless cleaning detergents (Schwartz, 1949):

1. Alkyl phenol and alkyl benzene sulfonates;

2. Sulfated fatty alcohols and sulfated oleic or ricinolieic acid;

3. Petroleum sulfonates;

4. Cetyl pyridinium bromide and other cationic agents;

5. Esters of long-chain fatty acids with low- molecular-weight hydroxycarboxylic acids, such as stearyl tartrate;

6. Oil-soluble nonionic agents, such as low HLB alcohol ethoxylates (HLB, hydrophilic-lipophilic balance of a surfactant).

As the above list indicates, very few oil-soluble surfactants fail to fit one of these classes.

A commercial waterless cleaning detergent may contain 40% to 90% active ingredients and be used at 0.5% to 4% in the cleaning solvent.

Quaternary ammonium compounds: surfactants with disinfecting properties. Among the broad spectrum of chemicals used in cleaning compositions, particular attention is paid to quaternary ammonium compounds. Quaternaries are unique surfactants that act as antistatic agents, detergents, oil-in-water emulsifiers, corrosion inhibitors, and lubricants. They can be either oil soluble, water soluble, or dispersible depending on the molecular weight and presence of fatty chains. While most commercial cationics contain one or more long alkyl chains, the most common types in waterless cleaning aids are based on diethylamine plus several moles of propylene oxide then quaternised with methyl chloride (Friedli, 2001).

Quaternary ammonium compounds are rather specific in their antimicrobial mechanism. Even very low concentrations cause damage to the cytoplasmic membrane due to perturbation of the bilayers by the molecules' alkyl chains (Wessels and Ingmer, 2013). QACs interfere with normal ammonium uptake (Sutterlin *et al*., 2008; Buffet-Bataillon *et al*., 2012). QACs irreversibly bind to the phospholipids and proteins of the membrane, thereby impairing permeability. Several active compounds have less inhibitory effect on *Pseudomonas* spp. than on *Bacillus* spp., due to the presence of lipoproteins and liposaccharides on the outer layer of peptidoglycane (Maris, 1995). Minimum inhibitory concentrations (MICs) of different QACs were recently summarised by Buffet-Bataillon *et al*. (2012).

McBain and coworkers (2004) examined QAC effects on bacterial community dynamics in a drain microcosm with mixed cultures. Increased susceptibility to QACs among some strains (e.g., belonging to genus *Pseudomonas sp.* and *Enterococcus saccharolyticus*) and decreased susceptibility among others (*Pseudomonas sp*., *Eubacterium sp*., *Chryseobacterium sp*., *Ralstonia sp*. and *Aranicola sp*.) was detected.

The probable production of QAC's per year in the EU is more than 1000 tons of pure compounds, several of the individual compounds being produced at more than 10 tons per year (Anonymous, 2013). It was previously reported that adaptation or resistance to QACs can develop (Reichel *et al*., 2014; Block, 1991; Boyce and Pittet, 2002).

ECOTOXICOLOGICAL IMPACT OF SURFACE CLEANING PREPARATIONS

Due to the enormous economic importance and massive worldwide use of surfactants and disinfectants, their environmental impact has to be evaluated and controlled. Especial attention is paid to ecological threat caused by water disinfection. In particular, the use of chlorine dioxide and ozone lead to bacterial mutagenicity (Monarco *et al*., 2000). A new class of disinfection byproducts (DBPs), i.e., halobenzoquinones (HBQs), which were observed to occur widely in treated drinking water and recreational water, were shown to be highly cytotoxic and potentially genotoxic and carcinogenic (Li *et al*., 2015). Hospital effluents possess environmental risks, since they are 5–15 more toxic than urban effluents (Panouillères *et al*., 2007). Panouillères *et al*., (2007) and Angerville *et al*., (2009) reported the effect of binary mixtures of detergents and disinfectants in different concentrations on the ecotoxicological status of hospital effluents. Acute toxicity tests performed on *Daphnia magna* with binary mixtures of i) sodium hypochlorite and three detergents or ii) peracetic acid and three detergents in different concentrations showed mostly antagonistic interactions (Panouillères *et al*., 2007; Angerville *et al*., (2009).

A complex preparation for waterless surface cleaning consisting of detergents, propanol, wax emulsion, mineral oil and quaternary ammonium compounds was found to exhibit strong disinfecting properties. Toxicity of this SCP for a battery of test organisms ranged as follows: crustaceans *Thamnocephalus platyurus* > algae *Selenastrum capricornutum* > Gram-positive bacteria *P. fluorescens* > higher plants *Triticum sp., Lepidium sativum* (Vecstaudza *et al*., 2015).

Data on toxicity of different types of detergents and disinfectants used for SCPs are shown in Table 1.

BIODEGRADABILITY OF SURFACE CLEANING PREPARATIONS

During biodegradation, microorganisms can either utilise surface active compounds as substrates for energy and nutrients or co-metabolise them by microbial metabolic reac-

Table 1

ECOTOXICOLOGICAL IMPACT OF SCPS AND THEIR ACTIVE INGREDIENTS

Active ingredient	Test organism/ Environment	Concentration	Results	Reference
Sodium hypochlorite	*Streptococcus pneumoniae and* coagulated blood	0.55%	Among different kinds of cleaning agents, sodium hypochlorite showed the best results in blood stems removing from hard surfaces and bacterial disinfection, but only when environment was not too wet	Gold and Hitchins, 2013
Quarternary ammonium compounds	*Pseudomonas fluorescens*	300 mg/L	MIC for growth inhibition during 24h batch cultivation	Davids *et al*., 2015
	Nitrifying bacteria	2 mg/L	First negative effects	IIvanković and Hrenović, 2010
	Algae *Dunaliella sp.*	0.79 mg/L	EC50 – after 24 h cultivation	
	Crustaceans *Daphnia magna*	0.38 mg/L	EC50 – after 24 h immobilisation	
Sodium dodecyl sulphate	*Acinetobacterjohnsonii* and *Oligotropha carboxidovorans*	0.2 mg/L 2 mg/L	Bacteria showed 50% and 20% viability during treatment with given concentrations of sodium dodecyl sulphate	
	Algae *Raphidocelis subcapitata*	36.58 mg/L	IC50 – cell density measurements after 72 h	
	Crustaceans *Artemia salina*	41.04 mg/L	LC50 – larvae mortality measurements after 24 h	
Alcohol ethoxylate	Crustaceans *Ceriodaphnia dubia*	0.39 mg/L	EC50 – after 48h immobilisation	
	Multiple algae species	0.030÷9.791mg/L	EC10*	
Nanosilver product-Nanocid® L2000	*Escherichia coli*, *Listeria monocytogenes*, *Salmonella typhimurium* and *Vibrio parahaemolyticus*	0.78÷100 µg/mL	MIC values showed that the MIC for all bacteria was 3.12 µg/mlL, but for *Listeria monocytogenes* it was 6.25 µg/ml. Based on contact time (for *Lysteria* it needs to be at least 7 hours), the MIC values for all bacteria was 6.25 µg/ml	Zarei *et al*., 2014
Alkyl ethoxysulphate	Crustaceans *Artemia franciscana*	23.92 mg/L	LC50** – Nauplii mortality after 72 h	Ivanković and Hrenović, 2010
	Algae *Pseudokirchneriella subcapitata and Raphidocelis subcapitata*	3.15 mg/L 2.18 mg/L	EC50* – Cell density 72 h IC50*** – Cell density 72 h	
	Different fish species	0.8 mg/L ÷250 mg/L	EC50	
Linear alkylbenzene sulphonic acid	Bacteria *Pseudomonas putida*	33.4 mg/L	EC50 – Growth inhibition after 16 h	
	Algae *Dunaliella sp.*	3.5 mg/L	EC50 – 24 h	
	Flathead minnow	0.63 mg/L ÷1.2 mg/L	First effect concentration on flathead minnow survival	Lewis, 1991
	Crustaceans *Daphnia magna*	1.7 mg/L÷3.4 mg/L	First effect concentration on *Daphnia magna* survival	
Nonionic alkyl ethoxylates	Crustaceans *Daphnia magna*	0.1 mg/L ÷1 mg/L	First effect concentration on *Daphnia magna* reproduction	

** LC50 – lethal concentration; *** IC50 – half maximal inhibitory concentration.

tions (Ying, 2006). Released into the environment, the biodegradability of QACs may be limited by their antimicrobial activity (Lucchesy *et al*., 2010).

The biodegradation process of SCPs is affected by physicochemical factors, e.g., solubility, concentration, structure of the target molecule, as well as conditions of media, e.g., dissolved oxygen, temperature, pH, light, nutrient concentration (Jurado *et al*., 2009, Jurado *et al*., 2013).

Although many bacteria are able to metabolise organic pollutants, a single bacterium does not possess the enzymatic capability to degrade all or even most of the organic compounds (Fritsche and Hofrichter, 2000). Biodegradation of organic pollutants by a consortium of microorganisms is more efficient. In biodegradation experiments with decyltrimethylammonium bromide (DTM), it was shown that *Xanthomonas sp.* cannot degrade DTM completely, and that the products of partial degradation would be available to

Table 2

BIODEGRADATION OF SCPs AND THEIR ACTIVE INGREDIENTS

Active ingredient	Inoculum	Results	Reference
Dimethysilanediol – monomer of polydimethylsiloxane	Bacterial and fungal cultures isolated from soils	*Fusarium oxysporum* converted 19% of [^{14}C]dimethylsilanediol to $^{14}CO_2$ after 240 days *Arthrobacter* sp. converted more than 10% of [^{14}C]dimethylsilanediol to $^{14}CO_2$ after 90 days	Sabourin *et al*., 1996
Organo-silicon compounds	*Pseudomonas fluorescens, P. putida*	Polydimethylsiloxane and other silicones can be transformed by a specific microflora	Wasserbauer and Zadak,1990
Quaternary ammonium salts	Activated sludge microorganisms	The resistance to biological degradation is to a small extent caused by increasing length of the alkyl chain. The resistance increases strongly with the number of long alkyl chains linked to the nitrogen atom	Van Ginkel, 1991
	Pseudomonas sp., Xanthomonas sp. from sewage sludge	Biodegradation of hexadecyltrimethylammonium bromide was evident at levels of 10 and 25 µg/mL The lack of activity on the hexadecyl compound at levels of 100 µg/ml probably is attributable to its toxicity. Decyltrimethylammonium bromide (DTM) was degraded faster	Dean-Raymond and Alexander, 1977
	Mixed population of microorganisms previously adapted to degrade the substrates	Three groups of the tested QAS differed in hydrophobic chain length or in hydrophilic properties. The degradation rate was influenced by the hydrocarbon chain length, the presence of aromatic or cyclic rings, and the occurrence of sulphur and oxygen atoms in the alkyl substituent. All tested QAS variants were biodegradable in an aquatic environment	Grabińska-Sota, 2011
	Pseudomonas putida	*P. putida* used tetradecyltrimethylammonium bromide (TTAB) as a sole carbon, nitrogen, and energy source. Specific variations in the content of phosphatidic acid, phosphatidylglycerol and cardiolipin in the membrane indicate that these phospholipids are involved in cellular responses to QACs, utilising principally phosphatidic acid to neutralise the high positive charge density given for the ammonium quaternary moiety from TTAB	Lucchesi *et al*., 2010
Alkylpolyglucosides sugar-based surfactants	*Pseudomonas putida*	For all the concentrations tested, the biodegradability proved lowest for the surfactant with the longest alkyl chain and greatest number of glucose units. The degree of biodegradation achieved is higher when the initial concentration of surfactant is lower. Lower concentrations, 15 mg/L and 25 mg/L, resulted in a biodegradation close to or above 90%	Jurado *et al*., 2013

support replication of *Pseudomonas sp.* so that the two organisms together would be able to grow on DTM (Dean-Raymond and Alexander, 1977).

Table 2 summarizes some results on biodegradation studies performed with different surfactants under different conditions.

CONCLUSIONS

Increased knowledge and better understanding of the antimicrobial capacity of disinfectants are essential to optimise sanitation procedures, to reduce costs, environment waste and to improve shelf life (Duong, 2005).

Special surfactants used in waterless cleaning vary widely. The literature on nonaqueous cleaning process is inadequate. More research is required to better understand the cleaning mechanism in solvent systems.

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BEZŪDENS TĪRĪŠANAS LĪDZEKĻI AR DEZINFICĒJOŠO IEDARBĪBU: EFEKTIVITĀTE UN APKĀRTĒJAS VIDES ASPEKTI

Arvien svarīgāka nozīme mūsdienās tiek veltīta dažādu līdzekļu izpētei, kurus izmanto medicīnas iestādēs, dzīvojamo un ražošanas telpu apkopē un dezinfekcijā. Rakstā apkopoti literatūrā atrodamie dati par preparātu specifiskām īpašībām tā sauktajā virsmas bezūdens tīrīšanā un dezinfekcijā. Sniegts apraksts par preparātu pielietošanas iespējām, ķīmisko sastāvu, testēšanas metodēm, kā arī preparātu ekotoksiskuma potenciālu un biodegradāciju. Preparāti savstarpēji atšķiras pēc ķīmiskā sastāva. To sastāvā ir iekļautas virsmas aktīvās vielas (VAV), šķīdinātāji, sāļi, mīkstinātāji, antistatiskas un dezinficējošas vielas. Īpaša uzmanība pievērsta četraizvietojamā amonija sāļiem, kuri veic VAV un dezinficējošās funkcijas. Bez tam šī vielu grupa piešķir preparātam antistatiskas, antiseptiskas, antikorozijas, emulģējošas u.c. īpašības. Lai novērtētu preparāta dezinficējošo iedarbību, jāņem vērā mikroorganismu sugas specifiskumu attiecība gan uz konkrēto antimikrobu aģentu, gan preparātu. Aktuāla problēma ir konkrēto mikroorganismu celmu rezistences veidošanās dezinfektantu iedarbībā. Šādā aspektā aktīvi tiek risināts jautājums par mikroorganismu iespējamo mehānismu veidošanos uz dezinfektantu un antibiotiku izturību. Daži autori uzskata, ka šie procesi ir savstarpēji saistīti. Svarīgi ņemt vērā arī izmantotās metodikas, testējot bezūdens tīrīšanas līdzekļus ar dezinficējošo iedarbību. Paralēli standarta metodēm svarīgi izmantot alternatīvu metodisko pieeju, modelējot bioplēves veidošanos uz specifiskas virsmas ar pārbaudāmo organismu pielietošanu, kuri izdalīti no konkrētās vides. Tā kā tīrīšanas līdzekļu ar augstāk aprakstītām īpašībām lietošana ir ļoti plaši sastopama, veidojas aktuāla vides piesārņošanas problēma ar preparātu ķīmiskiem komponentiem, kā arī ar to blakus produktiem. Autori apkopo datus par atsevišķu komponentu ekotoksiskumu un biodegradāciju. Rakstā minēti zinātniskie dati, kuri publicēti no 1977. līdz 2015. gadam, vienlaicīgi ietverot autoru pētījumu rezultātus.