# Biodegradation of typical laundry wastewater surfactants - a review

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Review

*The pollution of laundry wastewater is dependent on the origin of the linen, soil degree of the linen and the laundering process. It is caused by dissolved organic and inorganic substances, as well as sedimented and toxic substances. Washing detergents contain various chemicals used in great quantities whose influence on the environment is very important as they are transferred into wastewater treatment plants after use and are, therefore, also present in effluent, where they add their contribution to the total toxicity of the effluent. This review paper summarizes the findings of various literature and presents the biodegradation of most often used surfactants. In order to assess their environmental risks, we need to understand the distribution, behaviour and degradation of surfactants in the different parts of a wastewater treatment plant.*

***Key words:*** *aerobic biodegradation, anaerobic biodegradation, anoxic biodegradation, surfactants, toxicity*

## 1. Introduction

Different techniques are used for the treatment of wastewaters from laundry today; each with its own advantages and disadvantages. Several research reports have been published on the success of biological wastewater treatment of laundry wastewater [1-4]. In the past the most commons methods for treatment of laundry wastewater included sedimentation, flocculation, neutralisation, flotation. Lately more common methods include membrane filtration (micro-, ultra-, nanofiltration and reverse osmosis) as well as biological treatment (aerobic and aerobic) [5]. European Union and its member states have successively over the last three decades implemented European Union wide and national measures to ensure a sustainable water management process, an important outcome of which is the Water Framework Directive 2000/60/EC [6] of the European Commission which defines certain environmental targets that have to be carried out by all EU members.

In biological treatment systems, the biodegradable fraction of wastewater can be removed effectively, but its non-biodegradable fraction passes through the system unchanged. This review paper summarizes the findings of different researches and presents the possibility for biodegradation of the most often used anionic, nonionic, cationic and amphoteric surfactants. Its purpose is to help understand the behaviour of surfactants in different parts of wastewater treatment plants (anoxic, aerobic or anaerobic) and their possible presence in the effluent, where they can have a negative or even toxic effect on the environment.

## 2. Biological treatments

Degradation of surfactants through microbial activity is the primary transformation occurring in the environment. Biodegradation is an important process to treat surfactants in raw sewage in sewage treatment plants, and it also enhances the removal of these surfactants in the environment, thus reducing their impact on biota.

During biodegradation, microorganisms can either utilize surfactants as substrates for energy and nutrients or metabolize the surfactants by microbial metabolic reactions [7].
There are many chemical and environmental factors that affect the biodegradation of a surfactant in the environment. The most important influencing factors are chemical structure, and the physiochemical conditions of the environmental media [7]. Zhang and co-authors [8] reported that surfactant molecules with aromatic rings or secondary carbon chain structures have lower biodegradability in aerobic conditions. Increasing surfactant concentrations from sub- to supra critical micelle concentrations can significantly decrease biodegradation. This decrease may be attributed to the limited bioavailability of surfactants in the micelle phase as compared to the monomeric surfactants [8]. Surfactants are believed to act on the structure of the activated sludge flocks [9] and some of them are toxic to aquatic organisms. Surfactants containing a benzene ring are most likely to deteriorate wastewater treatment processes in the activated sludge system. The presence of the elevated amount of surfactants in wastewater can also decrease the affinity of substrate to biomass [10]. Different classes of surfactants have different degradation behaviour in the environment, therefore this review peper focuses on most widely used surfactants in industrial and domestic laundering of textiles.

### 2.1. Anionic surfactants

The anionic surfactant linear alkylbenzenesulfonates (LAS) can be degraded by a consortia of aerobic microorganisms and attached biofilms in the environment [11-13]. The primary degradation of LAS during the activated sludge treatment is more than 99 %, after an adequate period of adaptation (20 days) [14-15].Temmink and co-authors [16] reported that a large fraction (92–98 %) of LAS-$C_{12}$ in the municipal activated sludge plant was found to be adsorbed to the sludge and in general more than 99 % of the influent load of LAS-$C_{12}$ could be removed by biodegradation, indicating that not only dissolved but also adsorbed LAS-$C_{12}$ is readily available for biodegradation. However, LAS could be biodegraded primarily also by natural microbial flora [17-18]. Krueger and co-authors [19] reported the rate of LAS biodegradation increased with increasing the dissolved oxygen concentrations in the sewage-contaminated groundwater, but under low oxygen conditions (< 1 mg/L), only a fraction of the LAS mixture was biodegraded.
The inhibition of anaerobic digestion by LAS was studied by Mösche and Meyer [20] and by Hernandez and co-workers [21]. The latter reported that LAS exerts its toxic effect through an immediate inhibition and a subsequent further decrease of bacterial activity. Anionic surfactants besides their poor removal during anaerobic digestion may inhibit the process of hydrolysis [21]. Higher surfactant concentrations cause a faster decay of activity but, on the other hand, low LAS concentrations (< 3mg/L) might have a slightly positive effect on the bacteria [20].
Alkane sulphonates, alpha-olefin sulphonates, methyl ester sulphonates and sulphosuccinates (mono and dialkyl) can be biodegraded under aerobic conditions [22]. However, sulphosuccinates can also be partially (branched alkyl sulphosuccinates) or extensively (linear alkyl sulphosuccinates) mineralized in anaerobic digesters given that the ester linkage and its position with respect to the sulphonate group are essential for the degradation of the surfactant molecule under anaerobic conditions [22].
The mass flow and behaviour of secondary alkane sulphonates (SAS) and LAS during sewage treatment are similar in many regards. For example, the percentage masses of SAS and LAS transferred to sludge are similar, the more hydrophobic homologs and isomers of each surfactant are transferred to sludge preferentially, and both SAS and LAS are recalcitrant under anaerobic sludge digester conditions [23].
Alcohol sulphates or alkyl sulphates (AS) may interact in the wastewater treatment system in many ways. All interactions depend on the hydrophobic chain length. The most important interaction takes place with the cell membranes [24]. Depending on chain length and the composition of the cell membrane lipids and the chain length of the AS some surfactants will weaken these membranes more efficiently than other surfactants do. According to [25] the destruction of the cell membranes will cause an inhibition of the microbial activity and this inhibitory effect is permanent and not reversible by dilution [26]. The solubility of AS in water decreases with increasing hydrophobic chain length and this effect was considered to be one reason for the lower toxicity of long chain AS during anaerobic degradation [25].
Data on the biodegradation of anionic surfactants commonly used in industrial or domestic laundering of textiles is summarized in Tab.1.

### 2.2. Non-ionic surfactants

The presence of nonionic surfactants in wastewater at the concentration of 50 mg $L^{-1}$ in the influent causes a decrease of flocks size when the dilution rate is equal to or higher than 0.102 $h^{-1}$. The presence of both nonionic and anionic surfactants deteriorates the kinetics of biological wastewater treatment processes [27].
Biodegradable non-ionic surfactants such as alkyl polyglycosides are becomming more common [28].
Alcohol ethoxylates (AE orAEO) are believed to be a readily biodegradable and environmentally safe group of surfactants [17,29-30]. The results obtained showed that the biodegradability of alcohol ethoxylates depends on the molecular structure [31]. Mezzanotte and co-workers [31] observed

that the biodegradation of alcohol ethoxylates was affected by their affinity to water (the number of ethoxylate groups), length of the alkyl chain and molecular weight. High primary biodegradation (above 95 %) was also found for AEs in the continuous flow activated sludge test with a high concentration of metabolites free fatty alcohol (FFA) and poly(ethylene glycols) (PEG) [32-33]. Battersby and co-workers [34] tested the aerobic biodegradation of two AEs at 10 °C (as a winter temperature) and at 20 °C, and they observed more than 97 % of removal. In addition to aerobic biodegradation, rapid anaerobic biodegradation of AEs has also been observed [31, 35].

During sewage treatments nonylphenol (NP) and octylphenol (OP) can be efficiently removed from the sewage effluents through activated sludge treatments [13]. According to Isobe et al. [36] a greater removal for NP (93 % on average) than OP (84 % on average) was consistent with their partitioning behaviour to particles in primary and secondary effluents. Ma and co-workers [37] also reported that 71 % of NP and nonylphenol polyethoxylates (NPnEO) were removed by the sewage treatment process and the adsorption on sludge contributed to the removal of these compounds. Biodegradation of NPnEO s by denitrifying activated sludge was investigated by Lu and co-workers [38]. The results showed that NPnEOs were degraded readily (almost 90 %). Organic substance and initial concentration had great influence on biodegradation of NPnEOs in the denitrifying activated sludge process, while the influence of biodegradation intermediates such as NP could be neglected. Temperature and acclimation of biomass also has a strong influence on the biodegradation rate of NPnEOs, as it has been demonstrated that the biotransformation of these compounds is higher at a higher temperature [39] and the inhibition effect is smaller when the biomass is acclimatised [40]. Reports about the degradation of NPnEOs in anaerobic conditions are scarce. However, it appears that NP is the major product of degradation; it does not undergo further transformation and is adsorbed onto the sludge solids [41-42]. According to Luppi and co-workers [43] anaerobic degradation of NPnEO is strictly dependent on the presence of nitrate. There was no indication of biodegradation of NPnEO when nitrate was replaced by sulphate, bicarbonate or in electron acceptor-free controls. On the contrary Lu and co-workers [44] reported that anaerobic biodegradation of NPnEOs could be enhanced by adding sulphate or nitrate. Langford and co-workers [45] reported that sludge age is also important for biodegradation. Greater concentrations of activated sludge and, perhaps, greater species` diversity at a higher sludge age, enabled more rapid degradation of long chain NPnEOs. However, an accumulation of short chain compounds and NP was observed as their rate of formation exceeded their rate of degradation at all sludge ages and particularly at a higher sludge age due to rapid long chain compound degradation. Compared to anaerobic activated sludge treatment, denitrifying activated sludge treatment had a much higher removal efficiency of NPnEO contaminants [38].

Data on the biodegradation of non-ionic surfactants commonly used in industrial or domestic laundering of textiles is summarized in Tab.2.

### 2.3. Cationic surfactants

Tab.1 Biodegradation of anionic surfactants commonly used in industrial or domestic laundering of textiles

| Surfactant | Biodegradation | Conditions of successful biodegradation | References |
|---|---|---|---|
| Linear alkylbenzenesulfonates | aerobic | 20 days adaption period needed | [14,15] |
| Alkane-, alpha-olefin-, methyl ester sulphonates | aerobic | | [22] |
| Sulphosuccinates | aerobic | mono and dialkyl | [22] |
| | partly anaerobic | branched alkyl and extensively linear alkyl | [22] |
| Alkohol sulphates | partly anaerobic | short alkyl chain | [24] |

Tab.2 Biodegradation of non-ionic surfactants commonly used in industrial or domestic laundering of textiles

| Surfactant | Biodegradation | Conditions of successful biodegradation | References |
|---|---|---|---|
| Alcohol ethoxylates | aerobic | lower number of ethoxy groups, longer alkyl chain | [31] |
| | anaerobic | higher number of ethoxy groups, shorter alkyl chain | |
| Polyetoxylates | aerobic | / | [13] |
| | anaerobic | presence of nitrate | [43] |

Tab.3 Biodegradation of cationic surfactants commonly used in industrial or domestic laundering of textiles

| Surfactant | Biodegradation | Conditions of successful biodegradation | References |
|---|---|---|---|
| Quaternary ammonium based surfactants | aerobic | short alkyl chain | [48,51] |
| Diethyl ester dimethyl ammonium chloride | aerobic or anaerobic | none specified | [52] |

Tab.4 biodegradation of amphoteric surfactants commonly used in industrial or domestic laundering of textiles

| Surfactant | Biodegradation | Conditions successful biodegradation | References |
|---|---|---|---|
| Alkyl betaines | aerobic | none specified | [49, 53] |
| alkyl imidazole derivatives, alkylamido betaines | aerobic or anaerobic | none specified | |

Quaternary ammonium based surfactants (QASs) are commonly used in rinse cycles softener formulations. Under anaerobic conditions, some QACs showed poor primary biodegradation and no evidence of any extent of ultimate biodegradation [46-48]. However under aerobic conditions the cationic surfactants are considered biologically degradable, although the biodegradation for individual surfactants varies [49] and some of them can be toxic [50]. According to García and co-workers [51] an increase in the alkyl chain length or the substitution of a benzyl group for a methyl group reduces the aerobic biodegradation rate. Sütterlin and co-workers [48] investigated the biodegradability of QACs in the presence of different anionic surfactants. There was little biodegradability of the QACs as either single compounds or in the presence of organic counter ions. The biodegradability of the organic counter ions was lower in the presence of QACs as compared to the single substances. Primary elimination of the QACs by sorption took place. Cationic surfactants sorb strongly onto suspended particulates and sludge, which are predominantly negatively charged [48].

Due to the poor biodegradation kinetics of ditallow dimethyl ammonium chloride (DTDMAC) [47], diethyl ester dimethyl ammonium chloride (DEEDMAC) was introduced to replace it [52]. DEEDMAC differs structurally from DTDMAC by the inclusion of two ester linkages between the ethyl and tallow chains. These ester linkages allow DEEDMAC to be degraded rapidly and completely in standard laboratory screening tests and a range of environmental media such as sludge, soil and river water. DEEDMAC can be degraded completely under aerobic and anaerobic conditions, and it has a half-life of around 24 h in raw sewage. Therefore, removal of DEEDMAC during sewage treatment is greater than 99 % [52].

Data on the biodegradation of cationic surfactants commonly used in industrial or domestic laundering of textiles is summarized in Tab.3.

### 2.4. Amphoteric surfactants

Amphoteric surfactants form part of special surfactants available for formulators to improve or design new formulations in response to environmental, toxicity, safety and performance demands. Among the most important members of this family are the alkyl and alkylamido betaines and the alkyl imidazoline derivatives. Nevertheless, limited information on the ecological properties of amphoteric surfactants is available, Tab.4. As regards biodegradation, alkyl betaines, alkyl imidazoline derivatives and alkylamido betaines have been demonstrated to be readily biodegradable under aerobic conditions [49, 53]. Alkyl imidazoline derivatives and alkylamido betaines are also readily biodegradable under anaerobic conditions whereas alkyl betaines exhibit a negligible biotransformation [53].

## 3. Conclusion

The advantages of the physicochemical characteristics of surfactants used in laundering procedures have resulted in their industrial scale production and application all over the world. Besides the beneficial effects, they also show marked toxicity and can cause marked environmental pollution. Although most surfactants appear to be biodegradable, their complete mineralization relies on the activity and condition of the activated sludge. This review paper presents the degree of biodegradation for the most common used surfactants in industrial or domestic laundering of textiles in different conditions of biological treatment and, therefore, provides very useful information for the appropriate and successful treatment of the defined type of surfactants.

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