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Biodegradation of diclofenac by activated sludge and membrane bioreactor-A review

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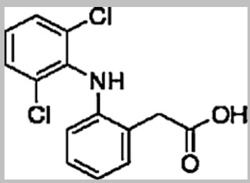
ABSTRACT

Abstract: *Diclofenac (DCF) is a pharmaceutical residue of therapeutic class of non-steroidal anti-inflammatory which is often detected in the wastewater treatment plants (influent and effluent) and surface waters. This review focuses its elimination by biodegradation with activated sludge (CAS) or bioreactor membrane (MBR) in which microorganisms plays a key role in the elimination of diclofenac and in which a lot of factors can affect the efficiency of the removal as physicochemical properties of diclofenac, sludge retention time (SRT), temperature, pH, redox conditions and sludge characteristics. The objective of this study was to describe a review of the literature by recent publications on the biodegradation of diclofenac. We inspect the performance of biodegradation using biological process technology by activated sludge and membrane bioreactor in the elimination of diclofenac.*

I. Introduction

Diclofenac (DCF), a polycyclic non-steroidal anti-inflammatory drug **[1- 3]** is one of the most extensively studied pharmaceuticals, for his potential toxic effects on no-target organisms **[4]**. Diclofenac was detected in wastewater **[5, 6]** in a number of countries such as China **[7, 8]**, Serbia **[9]**, Spain **[10, 11, and 12]**, Greece **[13, 14]**, Portugal **[15, 16]**, Brazil **[17]**, USA **[18]** and South Africa **[19]**. According to published documents, the concentrations of this drug varied in a range from 0.01 to 8.5µg/l **[20, 21]** and the rate of his elimination in sewage treatment plants is low (up to 40%) **[22, 23]. Table 1** presents physicochemical properties and molecular structure of Diclofenac. It was considered very toxic anti-inflammatory due to the death of birds recorded shortly after scanning the infected farm in India and Pakistan **[24, 25]**; he showed potential influence of endocrine disruption by delaying and reducing the success of fish eggs and hatching low concentrations damage the digestive organs **[25]**. Sewage treatment plays an important part in removing contaminants from reclaimed water but it's known that conventional wastewater treatment plants (WWTP) do not remove all the pollutants **[26],** especially the persistent polar pollutants due to their physicochemical properties such as diclofenac **[27, 28, 29]**. Most treatment methods of pharmaceuticals wastewater are biological processes **[30, 14 and 31]** in which microorganisms such as ammonia-oxidizing bacteria for the removal of ammonium **[32]**, Escherichia coli **[33]** and nitrite-oxidizing bacteria **[34]** are applied for removal of organic contaminants **[35, 25, and 36]**. Most WWTPs used are activated sludge processes **[37, 38]** and membrane bioreactors **[39, 40, and 24]**. Biodegradation by these two processes can be influenced by various factors namely; the physicochemical properties of diclofenac **[41]**, the biological operating conditions (sludge retention time, temperature, pH and redox conditions) **[42-44, 45]** and sludge characteristics **[46, 47]**. In this context, this study is a literature review of recent publications and studies to full and small scale on biodegradation of diclofenac to study the performance of activated sludge and membrane bioreactor in the elimination of diclofenac.

Table 1. *Main characteristics of diclofenac*

Structure a	Molecular weight (g/mol)a	Log K_{OW} a, b	pK_a a
	296.14	0.7-4.51	4.15

a : **[48]**, b: **[49]**

II. Biodegradation of diclofenac

Biodegradation is the process most usually used to eliminate the pharmaceutical residues persistent as the diclofenac **[50, 51 and 52, 53]**. This process decomposes and mineralizes these pollutants by bacteria or mushrooms in simpler compounds **[47, 54].** Diclofenac is a polar pharmaceutical compound mostly used as the sodium salt diclofenac-Na in human and veterinary medicine to reduce inflammation and pain **[5, 17 and 55]**. Many studies have evaluated the biodegradation of diclofenac by conventional activated sludge and membrane bioreactors.

2.1 Conventional activated sludge

Microbial degradation of diclofenac by activated sludge does not lead to its complete elimination but produces other metabolites which are also considered such as pollutants **[56]**, these authors investigated the repartition of diclofenac in activated sludge, they detected 7 diclofenac transformation products among them 1-(2,6-dichlorophenyl)-1,3-dihydro-2H-indol-2-one and 2,3-dichloro-N-(phenyl)aniline. The fungus Trametes versicolor can be utilized in activated sludge for the elimination of diclofenac **[57]**, in this study the removal rate was 64% which leads to a production of less toxic sludge, these authors concluded that fungal process can be an effective mean for biodegradation and diclofenac reduction in polluted waters. Activated sludge is the most used in the biological treatment and the rate of elimination of diclofenac by this process was insignificant and does not exceed 50 % **[6, 43, 58, 59, 60, 61, 62 and 63]**. Biodegradation of diclofenac can be estimated on measuring COD (Chemical Oxygen Demand) in polluted water and treated water **[64]**, **[60]** have illuminated this point and studied the elimination of diclofenac by activated sludge suspended in 2 reactors; ASR1 and ASR2. The removal rate was 48±19% and the concentrations of DCO decreased from 976±39 mg/l and 707±14 mg/l to 47±48 mg/l and 54±55 mg/l in ASR1 and ASR2, respectively. The hydrophobicity property defines the compound when the latter is insoluble in water **[47]**, **[65]** studied the effect of the molecular features of diclofenac in activated sludge in lab-scale experimental; they confirm that the functional groups can influenced the biodegradability. Among the biological process conditions; the sludge retention time SRT which may have an effect on the biodegradation and also affects microbial activity **[29]**, **[51]** evaluated the biodegradation of diclofenac in activated sludge of two wastewater treatment plants, Mamer and Boevange which differed in size, layout and sludge retention time (SRT). They observed that the biological degradation of diclofenac was significantly better in sludge from WWTP Mamer compared to sludge from WWTP Boevange and the removal rate decreases with increasing of SRT due to a lower active biomass presence. **[66]** reported the influence of SRT on diclofenac removal for more than seven months in denitrifying semi-continuous reactors operated at 10, 20, and 40 days by using two culture series (diclofenac free-control and diclofenac-acclimated); the results showed that the elimination rate for diclofenac-acclimated culture was lower than 15% a nitrate removal rate increased with increasing SRT but for diclofenac free-control this rate decreases with increasing SRT at 10, 20, 40 days with 48, 63, 79 mg NO^-_3-N/L day and 122, 55, 54 mg NO^-_3-N/L day, respectively. **[67]** investigated the biodegradation of diclofenac by nitrifying activated sludge and established the no removal of diclofenac with this process, these results are consistent with those of **[68]** who have studied the removal of diclofenac by nitrifying activated sludge in large scale (WWTP) and small scale (laboratory level) at a temperature of 12°C, the authors found that diclofenac is not biodegradable in large scale and the reaction rate constant (k′) and biodegradation constant (k_{biol}) were 1.7 d^{-1} and 0.6 l $g_{ss}^{-1}d^{-1}$, respectively in WWTP. In laboratory scale the biodegradation rate showed a difference between the reactors where with 10ug / l of diclofenac the biodegradation rate was 86% in the reactor 1, whereas in the other reactors was 90%. The authors explained this

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difference by the amount of active nitrifying bacteria in the reactors which was influenced by temperature. **[69]** studied the removal of diclofenac in two lab-scale conventional activated sludge reactors under nitrifying (aerobic) and denitrifying (anoxic) conditions for more than 1.5 years, they found that the removal by the biological treatment with nitrifying or denitrifying bacteria was not significant (<25%). Diclofenac in anoxic conditions (k_{biol}< 0.04 $Lg_{ss}^{-1}d^{-1}$) is persistent with respect to aerobic conditions (k_{biol}= 1.2 $Lg_{ss}^{-1}d^{-1}$) **[69]**. **[65]** found a similar partition coefficient (k_d) under aerobic and anoxic conditions (0.319 and 0.303 $L.gSS^{-1}$, respectively).

Sludge characteristics may influence the sorption diclofenac. These characteristics vary depending on the operation of treatment plants as activated sludge **[70]**. **[71]** Studied the biodegradability of the diclofenac on two types of sludge; sterilized and activated. Results of batch adsorption experiments via sterilized sludge showed that the removal efficiency was 40.1% at 6 hours and 19.7% for activated sludge where the contributions of sludge adsorption and biodegradation were 14.9% at 6 hours and 4.8%, respectively, The authors suggested that this difference in removal efficiency by sterilized and activated sludge is due probably by the sterilizing effect of the activated sludge which can be caused changes in the characteristics of the sludge and improves the elimination of diclofenac and also suggested that the structure of diclofenac (the electronic withdrawing groups) namely its functional groups (amine, halogen, and carboxylic groups) causes the resistance of diclofenac in its biodegradation.

2.2 Membranes bioreactors

Recently, the membrane bioreactor process has become the most effective process for the removal of diclofenac compared to activated sludge [72], where the authors have demonstrated that the removal rate of diclofenac by membrane bioreactor process was 65%. The molecular features of diclofenac can affect the rate of his elimination by membrane bioreactor on the scale of laboratory [73]. In this study, the authors observed a very low rate of elimination (17 %) and they justified this behavior of diclofenac in the MBR system by the presence of chlorine group and also the electron withdrawing functional groups generates an electron deficiency[74] investigated the degradation of diclofenac by a white-rot fungus-augmented membrane bioreactor (MBR) with and without addition of a redox mediator (1-hydroxy benzotriazole, HBT); the results showed that the addition of a redox mediator (1-hydroxy benzotriazole, HBT) improve the removal efficiency of diclofenac from 70% to 95%. On the other hand anaerobic MBR system has not shown an effective elimination rate for diclofenac (<10%) [75], these results are consistent with those of [76] these authors worked on a process of anaerobic / anoxic / aerobic membrane bioreactor in a large scale and observed removal rate below 20% for diclofenac. The full-scale MBR showed significantly better removal of hydrophilic compound (log D < 3) [77]; they explained the greater removal by the existence of preand post-anoxic tanks and the combination of aerobic zones with different levels of DO (Dissolved Oxygen) relative to a pre-anoxic and one aerobic tank in the pilot MBR. In contrast in another study [28] where they used an anaerobic system by anaerobic fluidized membrane bioreactor (AFMBR) using granular activated carbon (GAC) as carrier medium, 78% was the removal rate of diclofenac and the elimination rate of COD was 95% at total HRT of 5 h. It can be concluded that the use of GAC helps eliminate diclofenac in the anaerobic system. In non-sterile conditions studied by [72] the removal rate of diclofenac was 55% in an MBR system with a hydraulic retention time of two days [78] have shown that increasing the biomass concentration in the MBR system due to the long retention time favors the elimination of diclofenac and can be influenced by microbial activity according to [79]. These authors found that the reduction of microbial activity in the MBR system induces a decrease in the rate of elimination of diclofenac which indicates that the latter is eliminated by biological degradation and not by the sorption process only. [80] investigated the elimination of diclofenac by MBR under different temperature (10-45°C) in laboratory-scale and demonstrated that the temperature has an effect on the microbial activity. The results showed that the maximum and minimum removal rate were at 10 ° C and 20°C, respectively.

As regards the influence of pH, [81] explain the relationship between physicochemical properties of diclofenac and his efficiency removal by a laboratory scale MBR at mixed liquor pH values of 5, 6, 7, 8, and 9. At pH 5 the rate of removal was highest; this result is due to the physicochemical properties of this ionisable compound under acidic conditions wherein the pK_a value is 4.15 [82] so at pH 5 diclofenac is neutral which makes it a hydrophobic compound. The increase of the made pH decreases log D and consequently the hydrophobicity of diclofenac what makes decrease also the efficiency elimination. [83] confirmed the results found by [81] with their study where they assessed the biotransformation of diclofenac by laccase in an enzymatic membrane reactor, DCF biodegradation was better at acidic pH (3 and 4.5), and decreased with increasing pH.

3. Conclusion

The elimination of diclofenac was demonstrated to large and small scale with two process treatment of polluted water; biological process or biodegradation using conventional activated sludge (CAS) and membrane bioreactor (MBR) where the microorganisms are responsible for its degradation. Discussion of studies and publications used in this review demonstrated that biodegradation by membrane bioreactor is the most effective process for the removal of a resistant and persistent contaminant such as diclofenac with complete retention of suspended solids, thus reducing emissions to the dissolved fractions, but this removal may be influenced by various factors:

- Chemical structure where the presence of chlorine group and functional groups can influence the biodegradability of diclofenac.
- Sludge retention time (SRT) where the increase in this factor increasing the biomass concentration and therefore improves elimination of diclofenac.
- The temperature can affect the microbial activity in sludge and the maximum removal of diclofenac was seen at low temperatures.
- At acidic pH eliminating diclofenac was considered very favorable where the Hydrophobicity plays a very important role.
- Redox conditions: aerobic conditions in the elimination of diclofenac were better compared to the other conditions.

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