OC-12: Degradation of the Antiepileptic Drug Carbamazepin in Water by Hydrodynamic-Acoustic-Cavitation (HAC)

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The antiepileptic drug carbamazepine (CBZ) is a pharmaceutical with a widespread abundance in the aquatic environment due to anthropogenic effluents. The overall production is >1,000 tons p.a. worldwide (Zhang, 2008). Investigation of the effluent in different wastewater treatment plants (WWTPs) show that over 75% of the plants can stop the entry of CBZ only in the range of 0 - 10% (Zhang, 2008). The effect of low CBZ concentrations in the aquatic environment (1 - 50 µg L^{-1}) is discussed controversially, but ecotoxicological studies revealed reproduction toxicity, decreased enzymatic activity and bioaccumulation in different test organisms (Schwaiger at al., 2004; Ferrari et al., 2003; Vernouillet et al, 2010). Therefore, as a preventive step, efficient and economical technique for WWTPs is needed to stop the entry of pharmaceuticals in the aquatic environment.

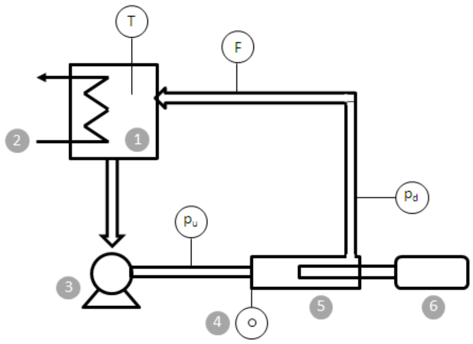

Figure 1 : Setup for creating Hydrodynamic-Acoustic-Cavitation (1 – reservoir, 2 – cooling, 3 – pump, 4 – orifice plate, 5- HAC reactor, 6 – ultrasound generator with sonotrode, measuring points for p_u – upstream pressure, p_d – downstream pressure, F – flow, T - temperature)

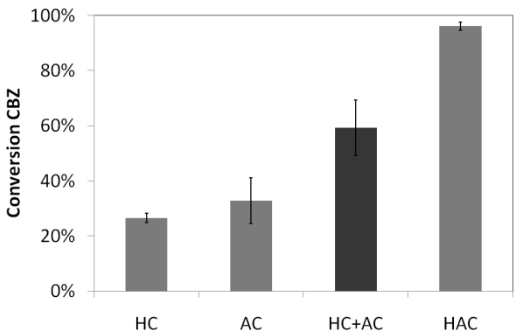

Figure 2: Comparison of different methods on the CBZ conversion (1 L, 5 µg L^{-1} CBZ, 25 °C, 15 min, 24 kHz, 125 µm)

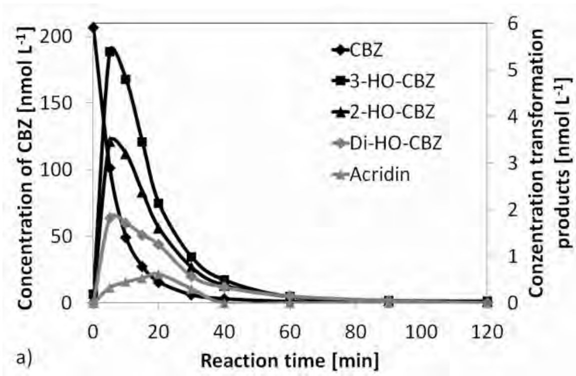

Figure 3: Time dependent development of the concentrations of CBZ and transformation products (HAC, 1 L, 48.8 µg L^{-1} = 206 nmol L^{-1}, 25 °C, 120 min, 24 kHz, 125 µm)

Cavitation, the formation, growth and subsequent collapse of gas- or vapour-filled bubbles in fluids (Suslick, 1989), is applied to solve the problem. In the collapse-phase high temperatures were generated in the bubbles (up to 20,000 K) leading to homolytic cleavage of water molecules to form highly reactive OH or OOH radicals for the degradation of contaminants in aqueous systems (Suslick and Flannigan, 2008). Cavitation can be generated either acoustic (ultrasound - AC) or hydrodynamic (HC) (Braeutigam et al., 2010). Both techniques have advantages and disadvantages, concerning the up-scaling, energy efficiency, produced temperatures and bubble density (Braeutigam et al., 2009). Therefore, a combination of both techniques in one reactor (Hydrodynamic-Acoustic-Cavitation - HAC) was introduced with a synergistic effect up to 19% based on the sum of the single methods (Franke et al., 2011).

HAC was applied for the degradation of CBZ in this study (Figure 1). Effects of the ultrasonic frequency, reaction temperature, CBZ concentration, restriction specific parameters, the distance between HC and AC, amplitude and pulse-ultrasound were investigated and used to optimize the system. Moreover, a kinetic study of CBZ and nine formed metabolites is performed. We were able to show that CBZ-degradation follows pseudo-first order kinetics to an extent of 27% by HC, 33% by AC and 97% by HAC within 15 min (Figure 2). A synergistic effect of 63% based on the sum of the single methods was found. After 60 min no metabolites were detectable (Figure 3).

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