## POLITECHNIKA ŚLĄSKA WYDZIAŁ INŻYNIERII ŚRODOWISKA I ENERGETYKI

ROZPRAWA DOKTORSKA

## Układ sekwencyjny fotokataliza - ciśnieniowa filtracja membranowa w pogłębionym oczyszczaniu odpływów z oczyszczalni komunalnych zawierających związki aktywne farmaceutycznie

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Gliwice 2016

## Summary

Presence of biologically active compounds in an aquatic environment, including pharmaceutical compounds may have a negative impact on living organisms. The main source of pharmaceutical micropollutants in the aquatic environment are wastewater effluents form conventional wastewater treatment plants, based on biological methods technology. Pharmaceutical compounds, due to their hardly-biodegradable nature, can only slightly be degraded by activated sludge microorganisms. Continuously increasing requirements for exploiters of wastewater treatment plants including the elimination of new groups of pollutants considered to be potentially dangerous, enforces the development of effective in-depth treatment methods.

The aim of research undertaken in the dissertation was to develop a complex method that ensures removal of compounds belonging to the group of non-steroidal anti-inflammatory drugs (ibuprofen, diclofenac) and psychotropic drugs (carbamazepine) from municipal wastewater after mechanical and biological treatment. The depth treatment process was carried out by photolysis, heterogeneous photocatalysis and pressure membrane process – nanofiltration.

The removal degree of pharmaceuticals in the photolysis process depends on the process time and the power of the applied UV lamp and exceeded 58%. The introduction of a photocatalyst to the reaction mixture (the process of heterogeneous photocatalysis) resulted in an intensification of the oxidation and reduction process of micropollutants.

It has been shown, that the efficiency of the process depends on: the type and dose of photocatalyst and the contact time with the reaction mixture prior to the irradiation process, the concentration of pharmaceutical compounds, pH level, oxygen concentration in the treated effluent and the volume of the reactor. For the most favorable conditions the elimination of diclofenac and ibuprofen exceeded 93% and 79% for carbamazepine. Moreover, the presence of inorganic compounds, including  $CO_3^{2-}$ ,  $HCO_3^-$ ,  $SO_4^{2-}$ ,  $Al^{3+}$  and  $Fe^{3+}$  ions inhibit oxidation mechanism of micropollutants. However the diverse physicochemical composition (presence of high molecular weight organic compounds) of the model and real effluent after biological treatment of wastewater intensified the decomposition of examined pharmaceutical compounds.

The processes of heterogeneous photocatalysis does not always allow for a complete mineralization of micropollution to  $CO_2$  and  $H_2O$ . Chromatographic analysis by GC-MS (EI) allowed to propose of pathways of photocatalytic oxidation of pharmaceutical

and toxicological analysis confirmed the toxic nature of the by-products generated during the decomposition of micropollutants.

The nanofiltration process of effluent after biological wastewater treatment allowed to decrease the concentration of micropollutants to a degree, which exceeded 90%. The determined value of the relative volumetric permeate flux as well as the performed analysis of the membrane topography indicated the occurrence of fouling phenomenon during membrane filtration, which resulted in the decrease in hydraulic performance of the membrane.

The sequential system, which integrates heterogeneous photocatalysis and nanofiltration process allowed for the complete elimination of both pharmaceutical compounds and metabolites generated during their natural and assisted decomposition. In these conditions also the occurrence of phenomenon of membrane fouling was minimized. The developed method, after the addition of economic analysis can provide a solution to the problem of increasing presence of pharmaceuticals micropollutants in the natural environment.