Non-Thermal Pulsed Plasma Discharge for Contaminants of Emerging Concern Removal in Water

Authors : Davide Palma, Dimitra Papagiannaki, Marco Minella, Manuel Lai, Rita Binetti, Claire Richard

Abstract : Modern analytical technologies allow us to detect water contaminants at trace and ultra-trace concentrations highlighting how a large number of organic compounds is not efficiently abated by most wastewater treatment facilities relying on biological processes; we usually refer to these micropollutants as contaminants of emerging concern (CECs). The availability of reliable end effective technologies, able to guarantee the high standards of water quality demanded by legislators worldwide, has therefore become a primary need. In this context, water plasma stands out among developing technologies as it is extremely effective in the abatement of numerous classes of pollutants, cost-effective, and environmentally friendly. In this work, a custom-built non-thermal pulsed plasma discharge generator was used to abate the concentration of selected CECs in the water samples. Samples were treated in a 50 mL pyrex reactor using two different types of plasma discharge occurring at the surface of the treated solution or, underwater, working with positive polarity. The distance between the tips of the electrodes determined where the discharge was formed: underwater when the distance was < 2mm, at the water surface when the distance was > 2 mm. Peak voltage was in the 100-130kV range with typical current values of 20-40 A. The duration of the pulse was 500 ns, and the frequency of discharge could be manually set between 5 and 45 Hz. Treatment of 100 µM diclofenac solution in MilliQ water, with a pulse frequency of 17Hz, revealed that surface discharge was more efficient in the degradation of diclofenac that was no longer detectable after 6 minutes of treatment. Over 30 minutes were required to obtain the same results with underwater discharge. These results are justified by the higher rate of H_2O_2 formation (21.80 $\mu molL^{-1}min^{-1}$ for surface discharge against 1.20 $\mu molL^{-1}min^{-1}$ for underwater discharge), larger discharge volume and UV light emission, high rate of ozone and NOx production (up to 800 and 1400 ppb respectively) observed when working with surface discharge. Then, the surface discharge was used for the treatment of the three selected perfluoroalkyl compounds, namely, perfluorooctanoic acid (PFOA), perfluorohexanoic acid (PFHxA), and pefluorooctanesulfonic acid (PFOS) both individually and in mixture, in ultrapure and groundwater matrices with initial concentration of 1 ppb. In both matrices, PFOS exhibited the best degradation reaching complete removal after 30 min of treatment (degradation rate 0.107 min^{-1} in ultrapure water and 0.0633 min^{-1} in groundwater), while the degradation rate of PFOA and PFHxA was slower of around 65% and 80%, respectively. Total nitrogen (TN) measurements revealed levels up to 45 $mgL^{-1}h^{-1}$ in water samples treated with surface discharge, while, in analogous samples treated with underwater discharge, TN increase was 5 to 10 times lower. These results can be explained by the significant NOx concentrations (over 1400 ppb) measured above functioning reactor operating with superficial discharge; rapid NOx hydrolysis led to nitrates accumulation in the solution explaining the observed evolution of TN values. Ionic chromatography measures confirmed that the vast majority of TN was under the form of nitrates. In conclusion, non-thermal pulsed plasma discharge, obtained with a custom-built generator, was proven to effectively degrade diclofenac in water matrices confirming the potential interest of this technology for wastewater treatment. The surface discharge was proven to be more effective in CECs removal due to the high rate of formation of H_2O_2, ozone, reactive radical species, and strong UV light emission. Furthermore, nitrates enriched water obtained after treatment could be an interesting added-value product to be used as fertilizer in agriculture. Acknowledgment: This project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie grant agreement No 765860.

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