PULSED CORONA DISCHARGE OXIDATION OF AQUEOUS DISSOLVED ORGANIC SUBSTANCES

F. SAPRYKIN[1], I. KORNEV[1], G. LOBANOVA[1], S. PREIS[2]

[1]Tomsk Polytechnic University, 2 Lenin Ave, Tomsk, 634028, Russia, kornev@tpu.ru
[2]Tallinn University of Technology, Ehitajate tee 5, Tallinn, 19086, Estonia

Atmospheric pressure gas-phase pulsed corona discharge (PCD) is a promising instrument in water oxidative treatment. Active oxidant species formed in air or oxygen discharge plasma, such as hydroxyl radicals (OH) and ozone (O_3), react with aqueous organic and inorganic dissolved impurities resulting in their decomposition and, also, water disinfection.

High reactivity of hydroxyl radicals and their short lifetime requires generation of the discharge plasma in close vicinity to target pollutants. Underwater and gliding surface discharges frequently used for this purpose concentrate energy in a few narrow channels resulting in radicals' recombination and low energy efficiency. The highest yield of active species and the treatment energy efficiency is achieved in gas-phase PCD. However, reactive species are largely produced in the gas phase, at a distance from the water surface, making the energy density in PCD with optimized mass transfer conditions in the plasma-water contact equipment the key factor for maximum oxidation efficiency.

The impact of process parameters on the oxidation efficiency of organic substances of various oxidation kinetics was studied. The discharge was formed in a wire-plate electrode system placed into a rectangular stainless-steel reactor sized 0.2×0.2×1.0 m. High-voltage wire electrodes were horizontally positioned between vertical grounded plates having the treated water flow perpendicular to wires. The distance between the wire and plate electrodes comprised 20 mm. Pulsed corona was energized by high-voltage positive polarity pulses with 200-300 ns duration, 20 kV amplitude and 0.3 J pulse energy. Water was dispersed in air flowing through a perforated plate: free-falling water droplets and jets passed through the discharge zone. The volume of treated samples was 20 L. In batch experiments, solutions were prepared in a storage tank, fed to the reactor for treatment, and recirculated after the treatment to the same tank. Solutions of humic acid sodium salt and oxalic acid were used in the experiments as target pollutants with relatively slow oxidation rates. Humic substances are widespread natural contaminants frequently found in natural waters. Slower reacting oxalate is a common byproduct of organic substances oxidation.

Experiments showed that parameters of the electrode system, such as the distance between the neighboring high voltage electrodes (10-30 mm) and the length of the high-voltage wire (8-25 m), did not have a noticeable influence on the oxidation rate of model substances, provided the discharge energy remained constant. With the same pulse energy, decreasing the wire electrode length from 25 to 8 m resulted in the volumetric energy density increased for more than three times. However, the discharge parameters, such as the energy, ozone yield and oxidation rate remained practically unchanged.

Increasing the energy consumption by changing the pulse repetition frequency from 200 to 860 pulses per second resulted in the increased oxidation rate at a reduced oxidation efficiency. The effect was better seen with slowly reacting oxalate, which was more efficiently oxidized at a lower pulse repetition rate, i.e. longer treatment time.

Accelerated water flow resulted in more efficient oxidation of oxalate: the oxidation rate increased by 30-40% when the flow rate changed from 0.5 to 2.4 $m^3 \cdot h^{-1}$, due to the increased water-air contact surface. However, improved contact between treated solutions and ozone present in the PCD reactor by its recirculating by means of a Venturi tube did not bring better oxidation.

An attempt was made to increase the residence time of water in the discharge zone by introducing polyethylene shavings into the inter-electrode area. This resulted in 30% lower oxidation efficiency of humic substances due to increased ohmic losses and discharge concentration visible by the discharge glow at fixed points of the plastic bed leading to high temperature in the discharge channels and lower yield of active species.

The most important parameter influencing the efficiency of organic substances oxidation appeared to be the discharge power, i.e. pulse repetition rate. Further research is needed in the mass transfer impact to oxidation efficiency.