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Treatment of Methylene Blue water solution by submerged pulse arc in multi-electrode reactor

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ABSTRACT

Pulsed submerged arc (SA) treatment of aqueous methylene blue (MB) solutions was studied in the reactor with multiple mobile iron (Fe) based electrodes. The discharges were carried out between these electrodes, which are repetitively brought into collision contact with fixed electrodes, connected to the power supply. The new reactor allows electrical discharges and MB decomposition in a large (600 ml) volume of liquid. The effects of alternating filtration, electrode type, arcing energy, vibration, aging of the solutions after arcing and the added H_2O_2 concentration on MB decomposition were studied. The ratio between the number of milliliters of the treated solution that reached complete removal of MB to the time required for this is greater by factor of 7.5 than this ratio for two electrode system obtained for the same SA conditions. The treatment efficiency in multi electrode reactor is explained by the numerous collisions of multiple electrodes leading to the formation of eroded nano-particles with the surface catalytically active towards MB oxidation.

1. Introduction

Treatment of wastewater is the inspiration for this study. Wastewater is an unwanted by-product from pharmaceutical factories, chemical processes, etc. It has been shown that plasma technologies can treat water using several mechanisms such as radical reactions, shock waves, ultra-violet radiation, ionic reactions, electron processes and thermal dissociation [1–6]. It is suspected that these factors, singularly or synergistically, may be responsible for concurrently oxidizing trace contaminates and disinfecting micro-organisms in water. The plasma can be located far from water or produced in the air adjacent to the water surface and can be submerged within the water [7–9]. When the plasma is submerged within the water, all of the radiation and active species produced are captured by the surrounding water. Thus this process might prove advantageous compared to others. This motivates investigation of the submerged arc (SA) discharge for several environmental applications, including drinking water and wastewater treatment [9]. The pulsed submerged (or electrohydraulic) arc discharge is a high current electrical discharge between two electrodes submerged in a liquid. It injects energy in the form of plasma which is surrounded by liquid [9–11]. Low-voltage ($\leq$100–200 V) pulsed SA discharges previously were used to decompose sulfadimethoxine and to decontaminate aqueous solutions containing other biological and chemical agents [12,13]. The SA in the two electrode reactors was also used to remove MB from aqueous solutions using carbon or iron electrodes [13,14]. Methylene Blue is a commercial dye which is used for various applications, mainly in medicine and the dye industry, however this dye is usually hard to remove by conventional means [15–18]. Its high solubility in water alongside easily monitored spectroscopy makes it a valuable tool for treatment efficiency estimation in regards to industrial waste water. The efficiency of MB removal using titanium electrodes with the addition of H_2O_2 was reported [19]. It was shown that MB decomposition continued when, after arcing, the treated solutions were "aged" in the dark for some time at room temperature [19]. The electrodes chosen are made of iron or steel, because of the possibility of producing Fenton's reagent (H_2O_2, Fe^{2+}), which very effectively oxidizes organic compounds [20–22], and because any iron micro- or nano-particles which are inevitably produced can be removed magnetically [23,24]. The cost of these electrodes is sufficiently low. SA is a relatively low cost method which might be invaluable for mass waste water treatment and reuse. Efficiency of SA treatment was proved for small doses of treated water, however no research justifying the concept of SA for a large scale process was reported.

The objectives of the present work were to: (1) determine the possibility of implementing the SA process in multi-electrode reactor which allows a treatment of the solution volumes more than 10 times larger than those treated in our previous works; (2) study

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the effects of the SA parameters and the H_2O_2 additives on the MB removal in the new reactor; (3) determine the efficiency of the SA treatment in multi-electrode reactor.

2. Experimental details

2.1. Arc treatment apparatus

The experimental system is presented in Fig. 1. The reactor consists of a stainless steel cylinder with the dimensions: 17 cm diameter, 1 cm outer shell thickness, and 14 cm height. The cylinder is standing upon a flat dielectric surface (bottom) wherein the stationary (fixed) electrodes are placed. The fixed electrodes are connected to the power supply. RC generator and industrial welding generator MOS210GEN were used separately as power suppliers. RC generator has three built-in capacitors of 5, 15 and 60 μF and provides output voltages of 32, 55 and 80 V. The currents of generator MOS210GEN did not exceed 20 A as higher currents could lead to welding of the movable electrodes. Usage of these power suppliers characterizing by the different pulse energy and frequency allows studying the effects of energy input to the system and of the reactor efficiency. Charge-discharge circuit of RC oscillator provides a pulse generation with a frequency of 100 Hz, while the welding generator provides frequency of 60 KHz. The reactor is secured in way that limits its horizontal movement (5–10 mm), but allows a small margin (2–5 mm) of vertical movement. Multiple mobile electrodes were placed at the bottom of the reactor. The reactor vibrations are created by transferring movements of the rotating cylinder mounted on the motor axis. "SHANDONG XIANGHEGROUP CO. LTD" Boshan micro motor ZYT-105 is operated by a separate power supply- "CARROLL&MEYNELL LTD" RPM. Rotating velocity is changed by applying voltage in the range of 0–260 V reaching 3600 RPM. Each one of the movable electrodes periodically contacts other movable electrodes and the fixed electrodes randomly forming a conducting path, and then quickly disengages. Current starts to flow when (or slightly before) the conducting path is formed, and the current continues flowing through an arc plasma which is created when the contact is broken. The arc parameters (pulse energy and frequency) in this multi-electrode system at the given voltage applied on the fixed electrodes are defined by quantity of vibrating multiple mobile electrodes in treated volume, by intensity of their moving and interaction. It must be noted that energy input to the system is determined by passing current through two phase system (treated liquid and multiple mobile electrodes) by two ways: arcs and leakage currents that may pass through the liquid under treatment in the intervals between discharges.

The arc voltage and current waveforms were recorded with a Tektronix TDS 2012 oscilloscope. Data obtained from oscilloscope were used to calculate such energy parameters of SA as pulse energy, and electricity needed for a volume of one cubic meter water treatment. Two kinds of multi-mobile electrodes were used: (type 1) steel hexagonal nuts, in the amount of 28 (weight 20.4 g, 1.4 cm thickness with 1.4 cm rib length and inner void radius of 0.7 cm) and (type 2) 15 mm diameter chrome steel AISI 52100G.040 balls, each ball weighing approximately 13.8 g. The balls were used in the amounts of 40, 60, 80 and 100. The SA parameters used in experiments are presented in Table 1 for a frequency of 2.4 Hz on ZYT-105 (if not specified otherwise).

2.2. Arc treatment procedure

The 10 mg/L MB solutions prepared using DI water (if not specified otherwise) were SA treated. The concentration of H_2O_2 in the MB solutions was varied in the range of 0–2 volumetric percents,

Table 1
System parameters used in experiments.

Power supplier	Electrode type	Electrode number	Pulse energy, mJ
RC-generator	1	28	40
	2	20, 40, 60, 80, 100	
	1	28	110
	2	20, 40, 60, 80, 100	
	1	28	217
	2	20, 40, 60, 80, 100	
Welding generator	1	28	0.7
	1	28	0.6 (1.5 Hz)
	2	40	0.7
	2	60	0.7
	2	80	1.2
	2	100	1.5

the treated volume was 600 ml (referred below as large volume) and the treatment time was varied in the range of 0–8 min.

The alternating filtration was used in a number of experiments. The suspensions formed after 2 min SA treatment were filtered through a "Whatman 595 1/2", in which the size of the pores is in the range of 4–7 μm. Then, the filtered solution was again arc treated. Filtration was repeated after every 2 min treatment. The total treatment time was 8 min.

After arcing, samples from the treated solutions were separated from the solutions in the reactor chamber in order to be "aged" at room temperature, and were examined at various aging times. The maximal aging time was one week.

For comparison, a number of experiments were conducted in two electrode system under the conditions used in our previous work [13] on which we refer below as on the standard conditions. The volume of the solutions treated in standard conditions was 40 ml and referred below as small volume. The system is based on manually induced contact between two iron electrodes submerged within liquid, that is placed inside a plastic cup. The two electrodes are contacted to the RC-generator; the applied frequency was 100 Hz.

2.3. Monitoring MB and H_2O_2 concentrations

Monitoring the MB and H_2O_2 concentrations in the SA treated solutions was conducted both immediately after treatment and for a long period after treatment (up to 30 h). In order to avoid decomposition of solutions under sunlight, samples from the treated solutions were stored in lidded plastic cups covered with aluminum foil paper to avoid penetration of visible and UV light.

The examination of the solutions was done by absorption spectroscopy using a stellarNet spectrophotometer, all measurements were done with a glass cuvette using the SpectraWiz computer program with temperature compensation and 194 ms integration time settings. The absorbance peak at 664 nm was analyzed for the MB monitoring. The absorbance peak values were used to calculate the MB removal based on the Beer-Lambert law. Background noise in spectra due to iron particles was eliminated by subtracting absorbance at 800 nm from peak absorbance.

Hydrogen peroxide concentrations were measured using the Ghormley method [25,26]; an absorbance of oxidized by hydrogen peroxide iodine was measured at 350 nm. Preliminary calibration was carried out for the added H_2O_2 concentrations in the range from 10 to 50 μM.

2.4. Characterization of eroded particles

The particles, eroded from the electrodes during SA treatment were characterized by X-Ray Diffraction (XRD) and by Scanning Electron Microscopy (ESEM).