



# Plasma-based water treatment: Conception and application of a new general principle for reactor design



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## HIGHLIGHTS

- Basic guidelines for plasma reactor design and optimization have been developed.
- Rhodamine B degradation depends on the area of the plasma–liquid interface.
- Reactor performance can be improved by generating foam on the liquid surface.

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## ABSTRACT

To improve the feasibility of plasma-based water treatment technology and develop basic guidelines for reactor design and optimization, a study was conducted to identify and characterize design parameters and physical phenomena that influence treatment efficiency. The first phase of the study established that the chemical reactions responsible for the degradation of organic solutes can be more accurately represented, mathematically, as heterogeneous reactions, rather than the common representation as homogeneous reactions. Using Rhodamine B as the model solute, the observed removal rate constant was found to be proportional to the area of the plasma–liquid interface. This observation supported the validity of the proposed heterogeneous rate equation and inspired the conception of a general design principle, which prescribes maximizing contact between the plasma and the treated solution. The second phase of the study involved the application of this design principle to create seven different “contact-oriented” reactors. The design parameters employed to increase contact included feeding liquid streams directly through the discharge region and generating a layer of foam on the liquid surface. The contact-oriented reactors validated their founding principle by achieving removal efficiencies up to 145 times that for the reference case (point-plate with discharge in liquid). The removal efficiencies attained in this work compare quite favorably with those achieved by other advanced oxidation processes for the degradation of Rhodamine B.

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## 1. Introduction

Advanced oxidation processes (AOPs) have increasingly been under investigation for their applications in the treatment of chemically and biologically contaminated water [1,2]. Some of these processes have reached a high level of development and have been commercialized, such as ultraviolet (UV)/hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) treatment and ozonation (O<sub>3</sub>) [3,4]. Pulsed electrical discharge plasma formed directly in or above water constitutes another, less-developed, AOP, but is receiving increased attention from researchers due to its unique treatment capabilities. Like

other AOPs, plasma-based water treatment (PWT) makes use of the highly oxidative hydroxyl (OH) radical to oxidize chemical contaminants, and thus does not yield the hazardous byproducts that may be produced by conventional chlorination processes [1,5]. PWT involves the generation of OH radicals in situ, and from the water itself; therefore, unlike most AOPs and conventional processes, little to no chemical additives are required. Additionally, plasma offers a broader range of chemical and physical treatment mechanisms. Besides producing several reactive chemical species (OH<sup>•</sup>, O<sup>•</sup>, H<sup>•</sup>, HO<sub>2</sub>, O<sub>2</sub><sup>•-</sup>, O<sub>3</sub>, H<sub>2</sub>O<sub>2</sub>, H<sub>2</sub>), the plasma channels formed directly in water can reach temperatures of up to several thousands of Kelvin and thermally degrade molecules, emit UV and visible light, and generate shockwaves capable of inducing cavitation [6,7]. While these features make plasma attractive as a

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stand-alone technology, they also open up the possibility of combining plasma with other AOPs. For example, the generation of UV light allows plasma to be effectively integrated with photocatalysis [8–10].

While the potential advantages of PWT are evident, the technology has not yet reached a high enough level of development to be used in practice. Some of the obstacles that have hindered progress in developing PWT systems are associated with the absence of general principles to guide the design of new reactors with higher efficiency.

The most expansive set of design criteria were offered by Malik in his review, where several different plasma reactors were compared in terms of treatment efficiency [11]. This review provided insight into which of the existing PWT systems performed best, and offered explanations for which process parameters contributed to higher treatment efficiencies. While there is much value in knowing which of the existing process parameters are most effective, it does not provide assistance to the development of new reactor designs.

The present study investigates how selected reactor design parameters affect removal efficiency of Rhodamine B (RhB) dye, and uses the resulting relationships to originate a more universal guideline for developing new reactors. In addition to providing insights into the fundamental nature of the overall mechanism responsible for organic solute degradation, this guideline is employed, to considerable benefit, for the development of several novel reactor types.

## 2. Experimental

The high voltage (HV) pulsed power supply used to generate the plasma was a custom built unit (Applied Physical Electronics LC, Spicewood, TX), with a general circuit diagram as shown in Fig. 1. The operational parameters used for this study were 20 kV discharges at 43 Hz, using a 0.94 nF load capacitor. The voltage and current in the plasma reactor were measured using a Tektronix P6015A high voltage probe and a Tektronix P6021 current probe connected to a Tektronix TDS 3032C oscilloscope. Examples of the voltage and current waveforms for discharges in both liquid and gas are shown in Fig. 2.

All reactors (Fig. 3) consisted of a 17.3 cm diameter glass vessel fitted with an airtight polymer cap, which was adapted to allow for sample extraction, solution recirculation, and integration of the electrodes. The reactors were operated in semi-batch mode, with liquid recirculating at 1.4 L/min. The liquid recirculation loop ensured thorough mixing and included a heat exchanger to keep the solution at 15 °C. The headspace was purged with argon at 2.1 L/min.

All reactors featured the point-plate electrode configuration, and differed in the size, shape, type and position of each electrode, as well as the mode by which the liquid feed stream entered the reactor. The “liquid discharge” reactor (Fig. 3(a)) contained a HV nickel chromium (NiCr) point electrode and a grounded stainless steel (S.S.) plate electrode both in the liquid phase with 1.5 cm

spacing. The “gas discharge” reactor (Fig. 3(b)) featured the grounded S.S. plate in the liquid and the HV NiCr point in the gas with 2.7 cm spacing (1.5 cm in liquid, 1.2 cm in gas). Four different sized S.S. plates were used: 0.64, 3.15, 4.75 and 7.60 cm.

For all of the “contact-oriented” reactors (Fig. 3(c)–(i)), the electrode positioning was similar to the gas discharge reactor, except a grounded aluminum ring (9.8 cm outer diameter, 6.0 cm inner diameter) was used in place of the grounded S.S. plate. In the turbulent jet reactor (Fig. 3(c)), the liquid feed was sprayed over the HV NiCr point with a 90° spray angle. The laminar jet reactor (Fig. 3(d)) was configured to introduce the liquid feed through a S.S. tube (0.32 cm inner diameter), which also served as the HV electrode. In the RVC HV reactor (Fig. 3(e)), the liquid feed passed through a reticulated vitreous carbon (RVC) plate, which served as the HV electrode. All of the “bubbling” reactors (Fig. 3(f)–(i)) were the same as the corresponding non-bubbling reactors, but involved bubbling argon (2.1 L/min) through a gas diffuser (9.5 cm diameter) centered beneath the hole in the ring electrode. The design characteristics of all the reactors are summarized in Table 1.

The treated solution was deionized water containing 7.5 mg/L (0.0157 mM) RhB with sodium chloride added to adjust the conductivity to 300 μS/cm. The pH was 5.4 for the untreated solution, and averaged 5.0 for the treated solution. The conductivity of the treated solution was 300–330 μS/cm. Due to variations in characteristics of the reactors, the solution volume was not the same for all experiments, ranging from 850 to 1350 mL; however, when appropriate, the reported results have been linearly scaled to represent a solution volume of 600 mL (chosen arbitrarily).

The concentration of the dye was determined spectrophotometrically (Shimadzu UV-1800) by measuring its absorbance at 554 nm. The concentration of H<sub>2</sub>O<sub>2</sub> was determined spectrophotometrically, using the reaction between H<sub>2</sub>O<sub>2</sub> and titanium sulfate and measuring the absorbance of the resulting yellow complex at 410 nm [12].

## 3. Results and discussion

### 3.1. Liquid and gas discharge reactors

The study began by using two of the most common reactor types, the point-plate reactor with discharges directly in liquid (Fig. 3(a)) and discharges in gas (Fig. 3(b)). As shown in Fig. 4, the performance of the liquid discharge reactor is poor, though the removal efficiency (discussed in Section 3.6) is of the same order of magnitude as those reported for this reactor type in other plasma studies [11]. The performance of the gas discharge reactor (Fig. 4) was significantly better, with an observed rate constant (discussed in the following section) approximately 16 times higher than that for the liquid discharge.

### 3.2. Solute degradation kinetics

To determine the reason behind the superior performance of the gas discharge compared to that of the liquid discharge, the nature of the overall degradation mechanism was evaluated. The shape of the concentration curves shown in Fig. 4 suggests first order kinetics, which is described by the following rate equation:

$$-\frac{dC_S}{dt} = k_{obs} \cdot C_S \quad (1)$$

where  $C_S$  (mg L<sup>-1</sup>) is the solute concentration (RhB in this case),  $t$  (min) is the treatment time, and  $k_{obs}$  (min<sup>-1</sup>) is the observed first-order rate constant. By integrating Eq. (1) and rearranging, an equation is obtained to calculate the observed rate constant from the concentration data:

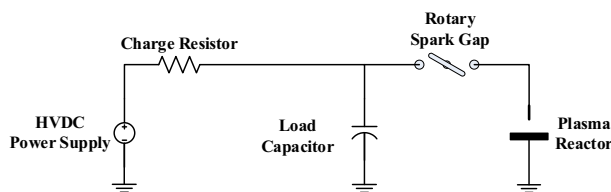


Fig. 1. Circuit diagram for pulsed power supply.

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