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Decomposition of phenol by hybrid gas/liquid electrical discharge reactors with zeolite catalysts

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Abstract

Application of hybrid gas/liquid electrical discharge reactors and a liquid phase direct electrical discharge reactor for degradation of phenol in the presence and absence of zeolites have been investigated. Hybrid gas/liquid electrical discharges involve simultaneous high voltage electrical discharges in water and in the gas phase above the water surface leading to the additional OH radicals in the liquid phase and ozone formation in the gas phase with subsequent dissolution into the liquid. The role of applied zeolites, namely NH_4ZSM5, FeZSM5 and HY, were also studied. Phenol degradation and production of primary phenol by-products, catechol and hydroquinone, during the treatment were monitored by HPLC measurements. The highest phenol removal results, 89.4–93.6%, were achieved by electrical discharge in combination with FeZSM5 in all three configurations of corona reactors. These results indicate that the Fenton reaction has significant influence on overall phenol removal efficiency in the electrical discharge/FeZSM5 system due to the additional OH radical formation from hydrogen peroxide generated by the water phase discharge.

Keywords: Gas/liquid corona; Phenol degradation; Zeolites; FeZSM5

1. Introduction

Advanced oxidation processes (AOPs) are based on the generation of highly reactive species, such as hydroxyl radicals that rapidly and non-selectively oxidize a broad range of organic pollutants [1–4]. Common AOPs involve Fenton and Fenton "like" processes, ozonation, photochemical and electrochemical oxidation, photolysis with H_2O_2 and O_3, high voltage electrical discharge (pulsed streamer corona and corona-like) processes, TiO_2 photocatalysis, radiolysis, wet oxidation, electron beams or γ-beams, and various combinations of these methods [5–7].

Pulsed corona, or corona-like, discharge processes utilize chemical radicals ($^{\bullet}OH$, $H^{\bullet}$, $O^{\bullet}$, $HO_2^{\bullet}$ and $O_2^{\bullet-}$) and highly reactive molecules (H_2O_2) produced from a high voltage pulsed electrical discharge that is sustained in an aqueous medium [8–13]. Hybrid gas/liquid electrical discharge reactors involve simultaneous high voltage electrical discharges in the liquid phase and in the gas phase above the liquid surface [14–16]. Two types of hybrid gas/liquid corona reactors, hybrid-series and hybrid-parallel corona reactors, have different electrode configurations and these differences affect the efficiency for organic compound degradation [17]. In both types of hybrid reactors the high voltage needle-point electrode is placed in the liquid phase as in the standard-reference corona reactor. The ground electrode, made from reticulated vitreous carbon (RVC), is submerged in the liquid in the standard-reference and hybrid-parallel case reactor, while in the hybrid-series reactor the RVC ground electrode was placed above the liquid surface. The parallel configuration of hybrid corona reactor employs an additional high voltage electrode made from RVC in the gas phase. These two reactor configurations, hybrid-series and hybrid-parallel, showed

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higher efficiency for degradation of organic compounds then the standard-reference corona reactor [15,17,18]. The hybrid reactors are more effective due to the formation of ozone in the gas phase which can either directly react with species in the liquid phase or, at high pH, can react with H_2O_2 formed in the liquid phase to produce OH radicals via peroxone chemistry, and the hybrid reactors may increase hydroxyl radical formation by the discharge at the gas–liquid interface [16]. Measurements of ozone produced in the gas phase in both hybrid reactors showed that approximately seven times more ozone is produced in the hybrid-parallel than in the hybrid-series corona reactor [14].

Phenol, one of the most abundant pollutants in industrial wastewater, was chosen as a model compound in this study due to extensive literature on phenol degradation by different types of advanced oxidation technologies [19–23], especially by applications including high voltage electrical discharge in water [15,16,24–28].

Zeolites were chosen as catalysts in this study due to demonstrated enhancement of organic dye compound degradation, particularly in the hybrid-parallel reactor, which was reported in previous work [17], and because it has been reported that highly oxidative media promotes oxidation reaction of organic molecules adsorbed on the surface of solid particles [28–31]. Additionally, the influence of different types of solid particles combined with corona discharge reactors for phenol degradation has been studied [25,28,31]. Furthermore, synthetic zeolites of ZSM5 and Y type showed rather good adsorption of phenol [32]. Zeolites may also act as ion exchangers because the loosely bound nature of the extra-framework metal ions allows for exchange of other types of metals when in aqueous solution [33–35]. The FeZSM5 zeolite is also of interest since the iron from the zeolites may react with H_2O_2 formed by the liquid phase electrical discharge to enhance organic compounds degradation as reported in the literature [36,37].

The aim of this study was to investigate the efficiency of the three different types of corona reactors including the standard-reference, hybrid-series and hybrid-parallel, in combination with the synthetic zeolites, NH_4ZSM5, FeZSM5 and HY, for phenol degradation.

2. Experimental

Three different types of pulsed corona reactors, namely standard-reference, hybrid-series and hybrid-parallel, were used for phenol degradation in this work (Fig. 1). The glass reactor vessel with a capacity of 1 l was used with the three different electrode configurations for each type of discharge. The reactor setup and electrode configurations were described in detail by Lukes et al. [14]. The pulse power supply was the same as in previous work, including pulse repetition frequency of 60 Hz and charging capacitance of 2 nF [3,8,14–18,27,28,31]. The applied peak voltage was 45 kV for each experiment which corresponds to the power input of approximately 60 W. Peak voltage, rise time and pulse width were measured by placing a Tektronix P6015A high voltage probe coupled to a Tektronix TDS 460 fast digital storage oscilloscope to the input of the pulsed power to the reactor. It should be noted that the measured peak voltage of 45 kV, rise time of the order of 20 ns and pulse width of approximately 3 μs FWHM are comparable with those given by Lukes et al. [14]. Moreover, the same waveform profiles were observed in experiments with and without zeolite addition into the reactors. Reactor current was measured with a P6021 Tektronix current probe. The corresponding energy per pulse of 1.06 (±0.05) J applied in all three reactor types was determined

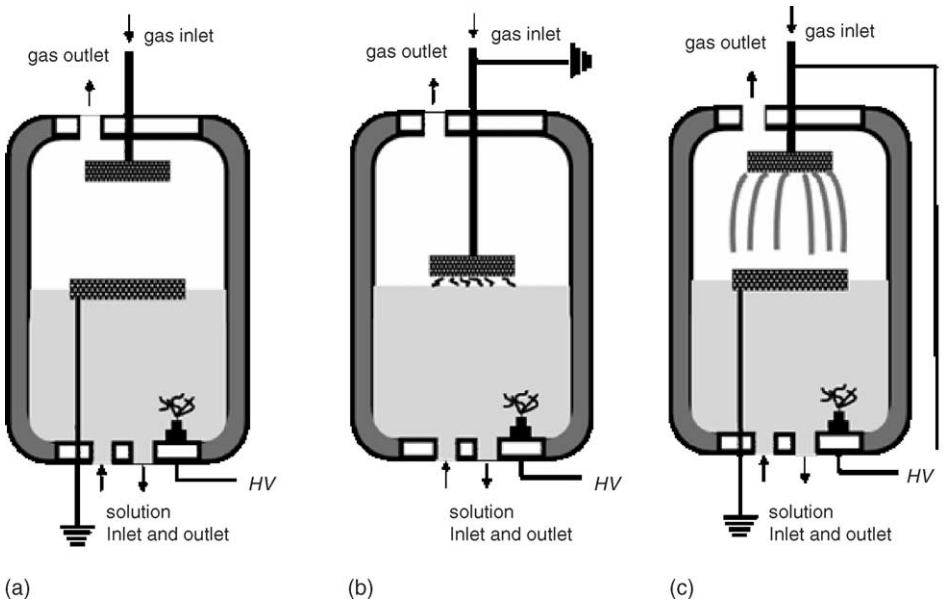

Fig. 1. Schematic of the three reactor configurations: (a) reference reactor, (b) series reactor and (c) parallel reactor.