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Microwave-assisted remediation of phenol wastewater on activated charcoal

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Abstract

In this paper, the effects of microwave dielectric heating in a multiphase gas–liquid–solid reactor are investigated. Experimental procedures are carried out in order to obtain significant arcing in the bed and control this cold plasma formation interpreted by Maxwell–Wagner effect. These procedures are tested on the catalytic wet air oxidation of phenol using activated carbon as catalyst. This study shows that arcing in the bed can only be obtained under conditions of partial wetting. Arcing leads to hot spots formation and thermal runaway if not early detected, but control is possible by acting on incident power. The low values of phenol conversions obtained are attributed to the very low oxygen partial pressure, experiments having been conducted at atmospheric pressure with air.

Keywords: Multiphase reactors; Microwave; Maxwell–Wagner effect; Adsorption; Oxidation; Activated carbon; Wastewater treatment

1. Introduction

Multiphase gas–liquid–solid reactors involving a solid catalyst are widely used in many industries and have been intensively studied but only a few studies, to our knowledge, have been concerned with gas–liquid–solid systems under microwave dielectric heating. For many gas–solid catalytic reaction systems, reaction rates are accelerated and product selectivity can be modified when microwave radiation is used rather than conventional heating (Bond et al., 1993; Gourari et al., 1992; Perry et al., 2002; Roussy et al., 1997; Zhang et al., 2001, 2003). In heterogeneous solid–liquid systems, many experiments have also shown significant improvement of the reaction rate under dielectric heating (Barnier et al., 1993; Hajek and Radoiu, 2000; Radoiu and Hajek, 2002). Whether this is due to pure thermal effects resulting from a selective dielectric heating or whether there are any specific microwave effects, is still the subject of many discussions.

Ability to convert electromagnetic energy into thermal energy depends on the dielectric properties of each material and can be described using complex permittivity ε. The real part ε' characterises a material ability to store charge, and ε'', the imaginary part, measure the heat related to electromagnetic losses in the material. Dielectric properties are dependent of composition, temperature and frequency. Thus, some solids are very efficient to convert electromagnetic energy into heat when gas and some liquids are totally transparent to microwaves. It is thus well established that the solid phase of a liquid–solid fixed bed can be selectively heated. Temperature profile measurements and modelling, at the surface of a liquid–solid fixed bed reactor under microwave heating, exhibits hot zones where catalytic reactions could be enhanced (Bonnet, 2003; Estel et al., 2003).

A few studies performed in gas–liquid–solid reactors under microwave heating are concerned with wastewater treatment. Shorrock et al. (2003) have developed a continuous process to adsorb and oxidatively decompose organic pollutants from aqueous effluent streams. Oxidation of Tributyl phosphate/water mixture is achieved in a gas–liquid fixed or fluidised bed of activated carbon, using air as gas

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reactant, under microwave heating. The reactor is operated in up-flow mode. Conversions are evaluated for several liquid and gas flowrates and for several masses of carbon. The decomposition of the contaminant is attributed to the formation of a high-energy plasma what is believed to be a Maxwell–Wagner effect. This effect is quite often mentioned in papers dealing with dielectric heating of granulated activated carbons and arcing phenomena or randomly fluctuating "hot spots" formation are described (Bond et al., 1994; Zhang et al., 2003). In fact, dielectric properties resulting from polarisation mechanisms, some interfacial polarisation occur for component mixtures and particularly between two phases. With active carbon which is an electrically conductor medium, interfacial charging phenomena appear and can lead to discharge phenomena coming with flashes. These high energy points can influence molecular behaviour and bring out some particular chemical phenomena like molecular break.

The objective of this work is to take benefits of the Maxwell–Wagner effect to improve the catalytic oxidation on activated carbon of phenolic wastewater. Catalytic wet air oxidation of phenolic effluents on activated carbon is usually performed in a gas–liquid fixed-bed reactor, at temperatures between 140 and 200 °C, and at pressures from 2 to 15 MPa (Fortuny et al., 1998; Polaert et al., 2002; Stüber et al., 2001). By operating the reaction under dielectric heating, the expected effect is a radical enhancement of the reaction due to interfacial polarisation and an oxidation under more moderate conditions.

Our investigations concern the two following points: (1) to obtain significant arcing in a gas–liquid catalytic bed and control the cold plasma formation in order to avoid thermal runaway; (2) to perform phenol oxidation in a trickle-bed reactor under arcing conditions.

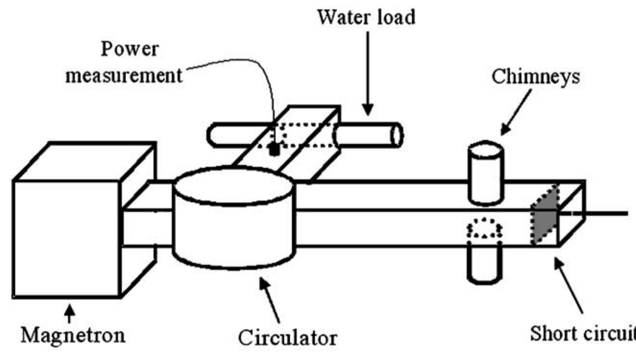

Fig. 1. Applicator design and specifications.

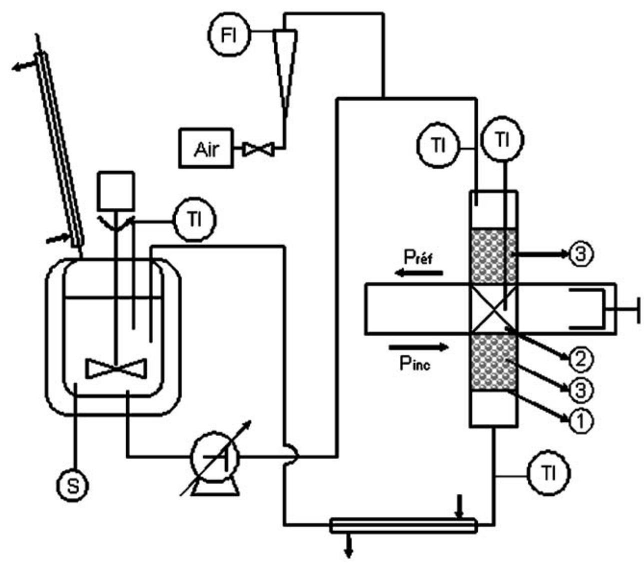

Fig. 2. Experimental set up.

2. Experimental

2.1. Materials

The commercial activated carbon used as catalyst was purchased from MERCK (Ref.1.02518.5000) in form of pellets. Prior to experiments, the activated carbon was sieved and only particles of 2.0–2.5 mm diameter were used.

Analytical grade phenol (Ph) was purchased from Aldrich and used without further purification. Bi-distilled water was used to prepare phenolic solutions and filtered compressed air used as gaseous reactant.

2.2. Experimental set up

Experiments were performed in a fixed-bed reactor introduced in the chimneys of a waveguide. As shown in Fig. 1, the microwave heating system consists of a magnetron working at 2.45 GHz with a maximal power of 1950 W. Microwaves are carried out in a WR 340 waveguide applicator for a resonant single mode TE_{013}. The applicator length is continuously adjusted to maintain reflected power at a minimum. The circulator protects the magnetron by conducting reflected power into a water load. Reflected and incident powers are measured and recorded.

As shown in Fig. 2, the fixed-bed reactor consists of a glass tube of 2 cm diameter equipped with a PTFE grid ① at the bottom. The activated carbon catalyst ② is placed between two 4 cm height beds of glass beads ③ ensuring a better distribution of the gas–liquid feed. The catalytic bed height is 4 cm and its position is adjusted so that all the activated carbon catalyst is in the irradiated zone. The reactor is introduced through the chimneys that prevent possible microwave leakage. Flow behaviour and sparkling inside the bed can be observed thanks to two small windows located at the irradiation zone level.

The liquid feed is stored in an agitated and temperature controlled vessel equipped with a condensation system connected to the vent line. The phenolic solution is pumped via