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Increase of acenaphthene content in creosote oil by hydrodynamic cavitation

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ABSTRACT

A hydrodynamic cavitation device was applied to increase the acenaphthene amount in creosote oil. In the presence of $FeSO_4$ and H_2O as an inducer and initiator, respectively, effects of H_2O amount, Fe^{2+} loading and pH of $FeSO_4$ solution were investigated by mean of the orthogonal experiment method, and effects of cavitating reaction temperature as well as the height of cavitator immersing in the feedstock by single factor method. The result indicated that the maximum increase was 7.06 percentage points under the optimum conditions as follows: H_2O amount 17.5% by the volume of feedstock, Fe^{2+} loading 0.3% by weight of feedstock, pH of $FeSO_4$ solution 1, the terminal temperature T of cavitating reaction 75 °C, and the immersing height of cavitator H 105 mm. Based on the change trend of components in creosote oil the reaction mechanism was derived, and the validity of derivation process was confirmed by the material balance.

1. Introduction

Creosote oil is a fraction of coal tar with the boiling range 230–300 °C [1], accounting for 9% of coal tar. It consists of more than 50 types of bicyclic and tricyclic aromatic hydrocarbons, mainly including naphthalene, 2-dimethyl naphthalene, acenaphthene, and dibenzofuran. As a component in creosote oil [2] acenaphthene has been widely used for the synthesis of drugs, pesticides, dyes, paints, resins, engineering plastics, and rubber stabilizers [1,3,4]. In generally acenaphthene is obtained from creosote oil by distillation, which has many disadvantages such as long process route, strict operation condition, and high energy consumption, attributed to its lower relative content, about 15%, in creosote oil.

Hydrodynamic cavitation is a phenomenon when liquid feedstock flows through a constriction such as orifice plate or/and venture, resulting in a sharp increase of the dynamic head at the expense of the hydrostatic head according to the principle of mechanical energy conservation. This leads to an ultra-high vacuum in the local area in which liquid feedstock vaporizes promptly and generates a large number of cavities. Subsequently, as the flow channel enlarges suddenly, a large amount of cavities collapse in a moment because of relative super-high-pressure outside, generating powerful energy, thus making the feedstock system to be in a supercritical condition and significantly reducing reaction energy barrier to generate shock waves and micro jets.

As reported, even in mild condition, the released energy is able to dissociate H_2O and dissolved O_2 in the system into highly active $H^•$, $HO^•$, and $O^•$ [5,6]. The produced shock waves and micro jets can enhance the collision among excited state feedstock molecules, thus lead to free radical reactions. Hence, the hydrodynamic cavitation, as a new intensification technology, has been applied to the process of organic wastewater treatment [7–11], tap water disinfection [12,13], hydrolysis [14], biodiesel synthesis [15–17], and so on.

A cavitation-oxidation technology developed in 1994 by a water treatment company of California quart in the United States has been used for the treatment of refractory organic wastewater, containing phenol, pentachlorophenol, benzene, toluene, ethylbenzene, dimethylbenzene, and cyanide. This way can improve the wastewater up to effluent standard [18].

Badve et al. [11] reported that the organics in wood processing wastewater can be degraded to CO_2 and H_2O by using a hydrodynamic cavitation device equipped with a rotary orifice plate at a rotating speed of 2200 rpm, where the chemical oxygen demand was lowered from 38,000 to 1672 mg/L.

Gogate et al. [19,20,21], have published several papers related to the intensification of chemical reactions by hydrodynamic cavitation. The paper [19] discussed the influence of the cavitation

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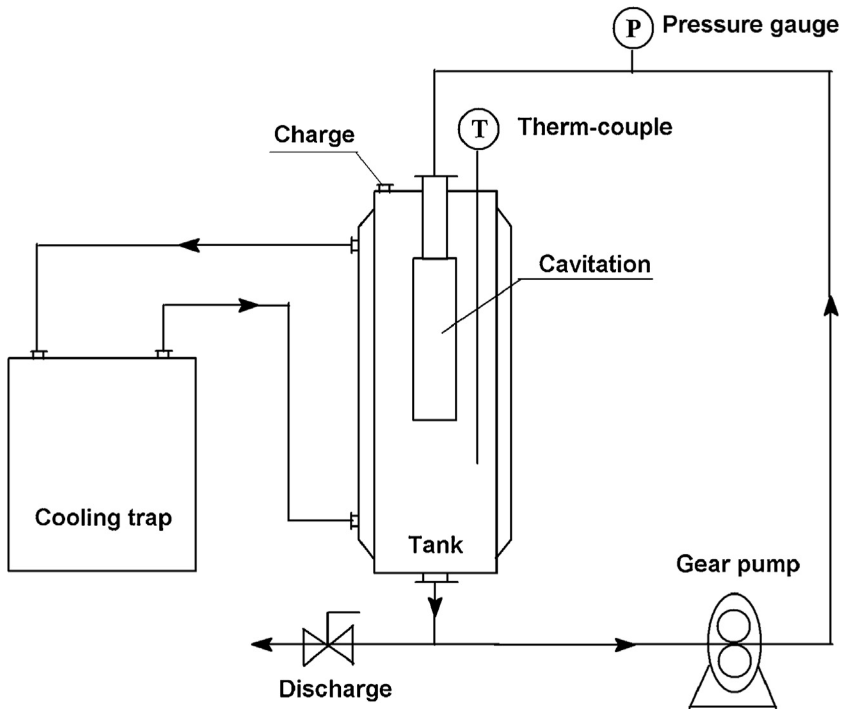

Fig. 1. Schematic representation of hydrodynamic cavitation reactor set up.

reaction on the degradation of water-borne guar gum in detail using cavitating devices such as orifice plate, circular venture and slot venture. Results indicated that with a slot venture, at polymer initial concentration c = 0.2% (W/V), inlet pressure P = 3 bar, and temperature of 47 °C, degradation degree of water-borne guar gum was 74.66%. If the amount of strong reducing agent KPS was added to 50 g L^{-1}, the degree reached up to 98.03%. The paper [20] investigated degradation of diclofenac sodium using a slot venture combined with ultraviolet irradiation and heterogeneous photocatalysis. Results indicated that degradation degree of diclofenac sodium was highly 94.78% at inlet pressure 3 bar, operating temperature 35 °C, reaction time 120 min, initial concentration 20 ppm and pH 4.0, and additions of photocatalyst TiO_2 0.2 g L^{-1} and oxidant H_2O_2 0.2 g L^{-1}. The paper [21] explained a hybrid reactor based on combined cavitation and ozonation: from concept to practical reality in depth. The principle involved the combination of hydrodynamic cavitation, acoustic cavitation, ozonation, and electrochemical oxidation/precipitation. When combined with different oxidation techniques, hydrodynamic cavitation increased effectively the generation of hydroxyl radicals, and made adequately the more contact of oxidizing species with pollutant to result in the pollutant degradation effectively.

This study focuses on increasing acenaphthene content in creosote oil based on the hydrodynamic cavitation reaction. In this paper the condition of cavitation reaction was investigated, reaction mechanism elucidated, material balance made, and cavitation number calculated. This process has not yet been reported.

2. Device and methods

2.1. Experimental device

Fig. 1 shows the schematic of experimental setup with circulation. It consist of a cavitator, vertical tank with the height and diameter (D) of 500 and 132 mm, gear pump, and pipe. The cavitator (ВКНТ-4.0/0.1, Russia) is tubular with the length (L) of 150 mm and diameter (d1) of 30 mm. As shown in Fig. 2 there are two orifice plates and a venture structure with a throat diameter (d2) of 18 mm in the cavitator. A pressure gauge was installed at upstream of the cavitator to monitor the stability, and a thermocouple was used to determine the feedstock temperature in the tank at any moment. To decrease feedstock heating rate and prolong the reaction time, the feedstock was cooled indirectly using a cooling trap with mixed liquor of glycol and water (1:1) at −15 °C.

2.2. Physicochemical properties of creosote oil

The oil sample was obtained from Xinlian coal chemical company in Xinjiang, China. The gas chromatography (GC) result was shown in Fig. 3. The boiling point and relative content of

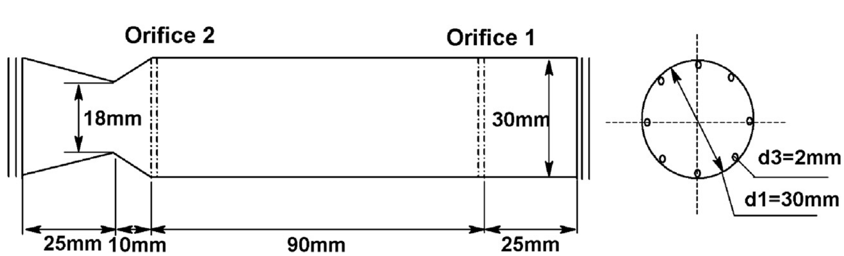

Fig. 2. Schematic representation of cavitation device.