



# Solubilization of polycyclic aromatic hydrocarbons (PAHs) with phenol in coking wastewater treatment system: Interaction and engineering significance

Qiaoping Kong<sup>a,b</sup>, Haizhen Wu<sup>a,\*</sup>, Lei Liu<sup>b</sup>, Fengzhen Zhang<sup>b</sup>, Sergei Preis<sup>c</sup>, Shuang Zhu<sup>d</sup>, Chaochai Wei<sup>b</sup>

<sup>a</sup> School of Biology and Biological Engineering, South China University of Technology, Guangzhou 510006, PR China

<sup>b</sup> School of Environment and Energy, South China University of Technology, Guangzhou 510006, PR China

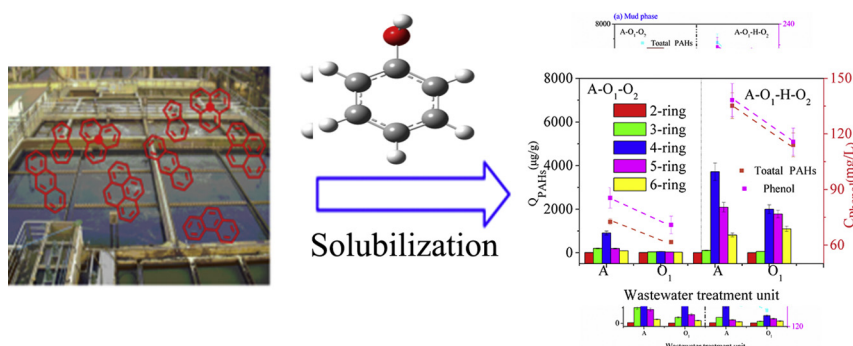
<sup>c</sup> Department of Materials and Environmental Technology, Tallinn University of Technology, Tallinn 19086, Estonia

<sup>d</sup> School of Biosciences and Biopharmaceutics, Guangdong Pharmaceutical University, Guangzhou 510006, PR China

## HIGHLIGHTS

- The distribution of PAHs and phenol in coking wastewater treatment with A-O<sub>1</sub>-O<sub>2</sub> and A-O<sub>1</sub>-H-O<sub>2</sub> reactor systems was studied.
- Phenol was found to be able to significantly improve the solubility of 16 selected PAHs.
- The value of  $\Delta\text{PAHs}/\Delta\text{phenol}$  was used to evaluate correlation between the biodegradation of phenol and PAHs.
- DFT calculation was used to investigate the binding energies between phenol and PAHs.

## GRAPHICAL ABSTRACT



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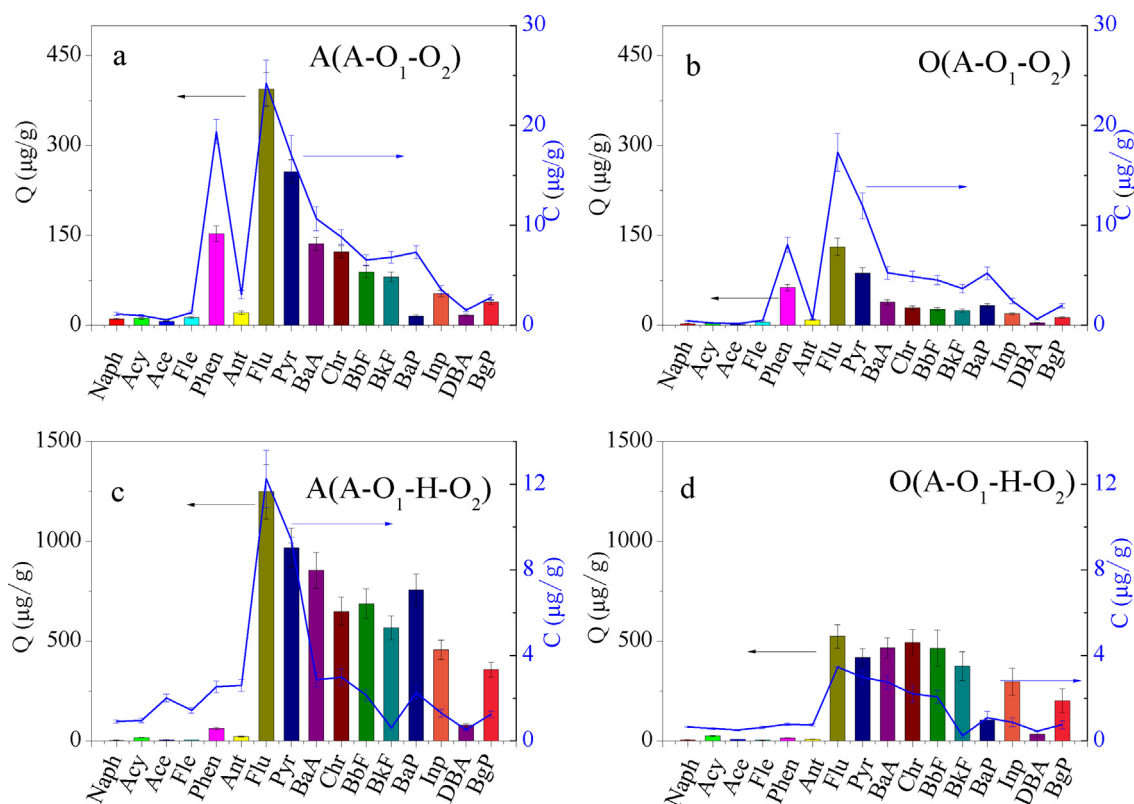
## ABSTRACT

Polycyclic aromatic hydrocarbons (PAHs) are accumulated in the sludge collected from the coking wastewater treatment. Phenol with its efficient degradation observed in biological treatment promotes the solubility of PAHs in aqueous phase. The interaction mechanism of phenol and PAHs in aqueous and sludge phases was systematically studied in two full-scale engineering projects composed of anaerobic-oxic-oxic (A-O<sub>1</sub>-O<sub>2</sub>) and anaerobic-oxic-hydrolytic-oxic (A-O<sub>1</sub>-H-O<sub>2</sub>) sequences. The results showed that reasonable use of phenol facilitates solubilization of PAHs alleviating their emission problems. The  $\Delta\text{PAHs}/\Delta\text{phenol}$  mass ratio in the sludge phase of A-O<sub>1</sub>-H-O<sub>2</sub> system (146.3) exceeded that in A-O<sub>1</sub>-O<sub>2</sub> one (63.80), exhibiting a good solubilization effect on PAHs with their more efficient degradation in the former. The full-scale observations were verified in laboratory solubilization experiments using phenanthrene (Phen), pyrene (Pyr) and benzo[a]pyrene (Bap) as the models of 3-, 4- and 5-ring PAHs, respectively. The binding energies of [phenol-PAHs] complexes were calculated using computational density functional theory showing consistency with the experimentally observed phenol-facilitated solubilization efficiencies in the row of Phen > Pyr > Bap. The results showed the fate and distribution of PAHs in coking wastewater treatment affected by the presence of phenol serving as a cost effective reagent for enhanced solubilization of PAHs from the coking wastewater sludge.

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\* Corresponding author.

E-mail address: [hzhwu2@scut.edu.cn](mailto:hzhwu2@scut.edu.cn) (H. Wu).



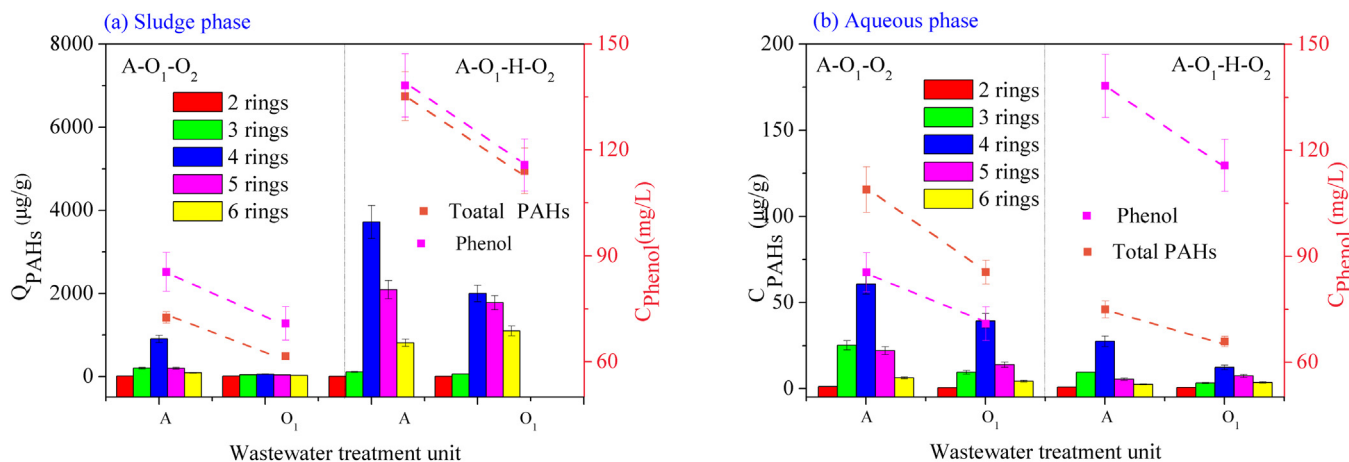
**Fig. 1.** Concentrations of PAHs in the sludge (Q) and aqueous (C) phases in the anoxic reactors A (a, c) and aerated reactors  $O_1$  (b, d) of A- $O_1$ - $O_2$  and A- $O_1$ -H- $O_2$  reactor systems: Q - bar graph; C - line.

## 1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) mostly originated from anthropogenic activities, such as coal mining, heat and power generation using fossil fuels, petroleum refining, coal and oil shale pyrolysis, and various chemicals production (Dai et al., 2007; Fatone et al., 2011; Zhang et al., 2012), are ubiquitous in industrial (Zhang et al., 2015) and municipal wastewaters (Liu et al., 2017). PAHs bring harm to the environment and human health due to their mutagenic and carcinogenic potential even at low concentrations (Ke et al., 2018). Hydrophobic PAHs are prone to be adsorbed by active sludge during wastewater biological treatment process (Wolejko et al., 2018) making their fate following a general trend of 'emission source-air-water-sludge-soil'. Coke production is considered to be one of the major sources of PAHs

contributing about 16% to the total PAHs emission in China (Zhang et al., 2012). High capability of coking wastewater sludge towards PAHs adsorption, 4–5 g/kg by dry weight, is determined by the factors including high PAHs concentrations in coking wastewater, long hydraulic retention (HRT) and sludge retention (SRT) times.

For the removal of PAHs in wastewater sludge, a wide range of methods including combustion (Moore, 2006; Wen et al., 2016), pyrolysis (Stark and Ghoniem, 2017), ozonation (Carrere et al., 2006), Fenton oxidation (Chen et al., 2015; Usman et al., 2016),  $H_2O_2$  oxidation (Malakahmad and Ho, 2017) and ultrasound processing (Sponza and Oztekin, 2011; Ke et al., 2018) were applied, depending on inorganic and organic contents in the wastewater sludge. In a viewpoint of practical application, however, the utilization of those methods is challenged by high loadings of PAHs and other organic pollutants. The



**Fig. 2.** Distribution of phenol and PAHs of various structures in aqueous and sludge phases in A and  $O_1$  reactors of the A- $O_1$ - $O_2$  and A- $O_1$ -H- $O_2$  systems.

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