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# DEGRADATION OF AROMATIC COMPOUNDS USING MOVING BED BIOFILM REACTORS

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## ABSTRACT

For biological treatment of water, there are many different biofilm systems in use. Examples of them are trickling filters, rotating biological contactors, fixed media submerged biofilters, granular media biofilters and fluidized bed reactors. They all have their advantages and disadvantages. Hence, the Moving Bed Biofilm Reactor process was developed in Norway in the late 1980s and early 1990s to adopt the best features of the activated sludge process as well as those of the biofilter processes, without including the worst. Two cylindrical moving bed biofilm reactors were used in this study working in upflow stream conditions. Experiments have been done in aerobic batch flow regime. Laboratory experiments were conducted at room temperature (23–28°C) and synthetic wastewater comprising a composition of phenol and hydroquinone in each reactor as the main organic constituents, plus balanced nutrients and alkalinity were used to feed the reactor. The ratio of influent to effluent COD was determined at different retention times. The results indicated that the removal efficiency of each selected compound is affected by the detention time. At low phenol and hydroquinone concentration (from 700 to 1000 mg/L) maximum removal efficiency (over 80 %) was obtained. By further increasing in COD loading rate up to 3000 mg/L, a decrease in COD removal rate was occurred. In the reactor containing pyrogallol in COD of 1500 mg/L, the removal rate decreased to 10 percent because of its toxicity for microorganisms.

**Key words:** Hydroquinone, Moving Bed Biofilm reactor, phenol, pyrogallol.

## INTRODUCTION

The removal of toxic phenolic compounds from industrial wastewater is an important issue to be addressed. Phenolic compounds such as phenol, hydroquinone and pyrogallol are used in different industries which end into their wastewaters. They are considered as priority pollutants because of their high toxicity at low concentrations. Phenol is an important raw material in petrochemical, pharmaceutical, plastic, pesticide production industries. Hydroquinone and its derivations is another phenolic compound used in photographic applications, rubber industry, monomer inhibitors, dyes and pigments, antioxidants agricultural chemicals, and other diverse and special applications. The largest demand for hydroquinone is as a photographic developer, principally for black and white film, lithography, photochemical machining, microfilm, and X-ray film (Elves *et al.*, 1989a). Pyrogallol is also the other phenolic compound that used in photography, lithography and hair dyes production. It is also used as an antioxidant and stabilizer. The use of pyrogallol in the field of cosmetics and medicines is currently declining because of pronounced toxicity (Elves *et al.*, 1989b). The presence of these pollutants in water and soil has become significant problems. Effective methods for the removal or treatment of them need to be pursued.

Many efforts have been made for the biological treatments of wastewater rich in phenolic compounds. Common commercial wastewater treatment methods utilize the combination of physico-chemical and biological treatment. Both chemical and biological processes were used for many years to treat phenolic wastewater.

Activated sludge, fluidized and packed bed reactors and recently moving bed biofilm reactors (MBBR) were studied as biological treatment

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processes (Vinod *et al.*, 2006; Murugesan *et al.*, 2005; Hosseini *et al.*, 2005; Andreottola *et al.*, 2000). Chemical processes like advanced oxidation methods by ozone, hydrogen peroxide and fenton like process (Ri-Sheng *et al.*, 2006; Wu1 *et al.*, 2002) and reduction by electrode reactor (Xiong *et al.*, 2003) were also studied for degradation of phenol. Laboratory experiments are done on adsorption of phenol on carbonaceous adsorbents as an effective advanced process to treatment phenolic wastewater (Ahmaruzzaman *et al.*, 2005). The moving bed biofilm reactor process (European Patent No.0, 575,314, US Patent no. 5,458,779) was developed in Norway in the late 1980s and early 1990s. In 1988, the Norwegian State Pollution Control Authority made recommendation on design of small wastewater plant (Rusten *et al.*, 1997). These recommendations include design of biological/chemical treatment plants based on low loaded biofilm process with a large tank (sludge separator) serving both as pretreatment unit, sludge holding and equalization tank. A Norwegian company (Kaldnes Miljøtekonlogy A/S), which was developing the so-called MBBR at the time initiated construction of small treatment plants according to these recommendations.

MBBR has been a commercial success. There are presently more than 400 large-scale wastewater treatment plants based on this process in operation in 22 different countries all over the world. In addition there are several hundreds small, on-site treatment units based on the MBBR mostly in Germany. More than 50 MBBR plants are in operation at commercial fish farms, in addition to several hundred small MBBR systems for ornamental fish. MBBR is performed in biofilm system with relatively large (0.1 to 5 cm) carriers. These are mixed with the wastewater and suspended in the reactor by the turbulence. The system is located somewhere between an activated sludge and a fixed-bed biofilm system (Maurer *et al.*, 2001). One important advantage of the moving bed biofilm reactor is that the filling fraction of biofilm carriers in the reactor may be subject to preferences. In order to be able to move the carrier suspension freely, it is recommended that filling fractions should be below 70% (Rusten *et al.*, 2006). One may, however, use as much as needed below this. A number of different carriers have been developed. Those designed and developed in Norway, are two variants called Kaldnes® and Natrix® (Maurer *et al.*, 2001). These carriers are small plastic tubes (1 to 5 cm diameter or length) made from polyethylene with density close to 1 g/cm$^3$. The inner part of the tubes is divided into several sectors to increase the total biofilm surface. This system is successfully used for treating of high-strength industrial wastewater. Also other kinds of carriers with differences in size and shape and, therefore, in different specific surface areas have been used. Investigations on the shape and size effect of carrier made it clear that the key factor in the design of a moving bed biofilm process for organic matter removal is the effective surface area where biomass may grow. The size and shape of carrier may have an influence on this effective area. The design of process should be based on organic surface area removal rate (Ødegaard *et al.*, 2000).

## MATERIALS AND METHODS

Three applied cylindrical MBBR reactors (Fig. 1) were made of Plexiglas. Each reactor had an internal diameter of 10 cm, a height of 70 cm and wall thickness of 4 mm and five sampling ports. The effective depth of wastewater in each reactor was 60 cm (70% of reactor volume) filled with plastic floating biofilm carrier (Fig. 2) described in Table 1. Batch reactors were working in upflow stream conditions in room temperature (22-26 $^{o}$C). The circulation of the biofilm carriers inside the reactors was caused by aeration. In order to keep the carriers in the reactors, a sieve (with 5 mm opening) was placed at the outlet of the reactors. Filling ratio of plastic elements in the reactors is important due to the amount of biomass, which can be supported by carriers. The reactors used were filled about 70% (recommended percentage volumetric filling of plastic elements in empty reactor). The synthesized wastewater has been prepared using phenol, hydroquinone and pyrogallol which were supplied by Merck, Germany.

In order to have C/N/P= 100/5/1 and alkalinity, necessary nutrients (urea, $KH_2PO_4$, $K_2HPO_4$) were used to feed the reactor. The parameters of pH, COD (soluble COD filtered through Vattman

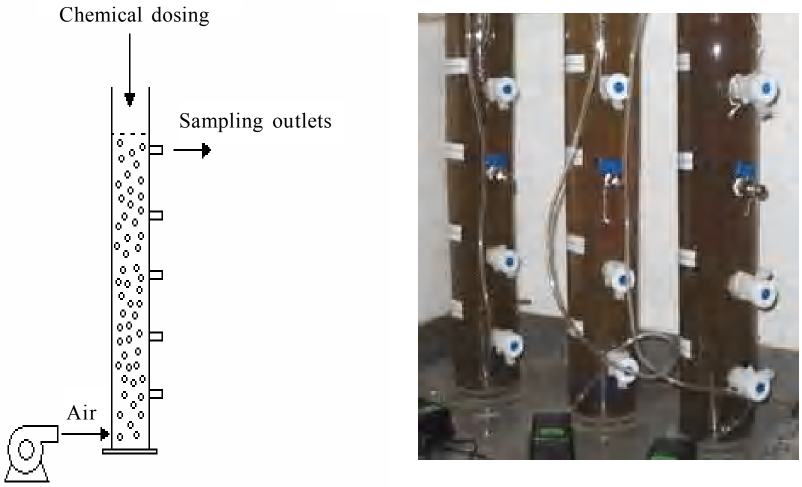

Fig. 1: Schematic diagram of the MBBR and the reactors used in the study

paper No.42) and dissolved oxygen (DO) were measured daily. TSS, mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) were measured on alternative days. Volatile Fatty Acids (VFA) and alkalinity were controlled weekly. Microscopic investigation was done regularly. All analytical tests were done as outlined in the Standard Methods Handbook (APHA, 1998).

Fig. 2: Biofilm carriers used in experiments

Table 1: Characteristic data for the carriers

| Material | Polyethylene |
|---|---|
| Density (g/cm$^3$) | 0.95 |
| Shape | Corrugated cylinder |
| Length (mm) | 120-150 |
| Diameter (mm) | 120 |
| Specific surface (m$^2$/m$^3$) | 120 |
| Filling ratio (%) | 70 |

## RESULTS

After 20 days of starting up the reactor with sludge obtained from Ekbatan wastewater treatment plant as seed, solution of glucose and wastewater compound with COD of 1000 mg/L with different concentrations were used. In the adaptation period, the amount of Organic Loading Rate (OLR) was being increased stepwise within 60 days. In the beginning, the COD removal rate was very low but after 4 months of the study, it reached to 50% that are more discussed in the following paragraphs.

### *Effect of detention time on COD removal rate*

The effect of different retention times was studied for each reactor. COD removal efficiency has measured after 24, 48 and 72 hr for every step increase in COD. At low phenol and hydroquinone concentration (from 700 to 1000 mg/L) maximum efficiency was obtained. By further increasing in COD loading up to 3000 mg/L, a decrease in COD removal rate was occurred.

Same behavior in reactor containing hydroquinone was observed. However, decrease in COD removal rate in hydroquinone reactor was slighter. It seems that further increasing in concentration of hydroquinone and especially in phenol will eventually cause the physiological parameters decrease and the substrate inhibition occur. Figs. 3 to 5 indicate the removal rate for different COD and detention times for phenol, hydroquinone and pyrogallol, respectively.

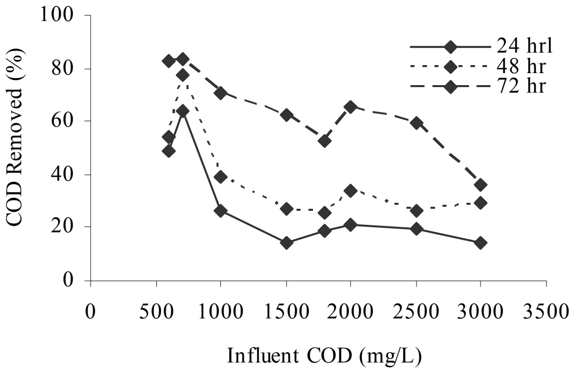

Fig. 3: Variation of organic loading rate and COD removal for phenol wastewater in different detention times

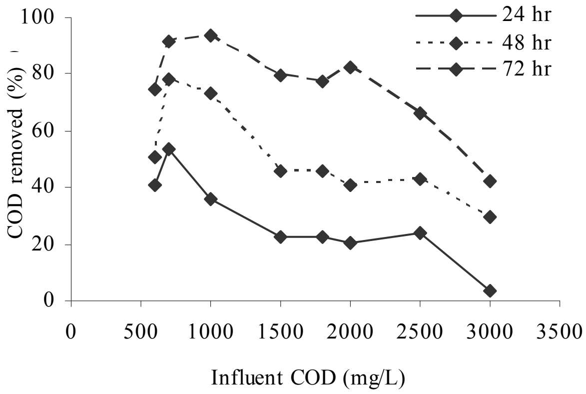

Fig. 4: Variation of organic loading rate and COD removal for hydroquinone wastewater in different detention times

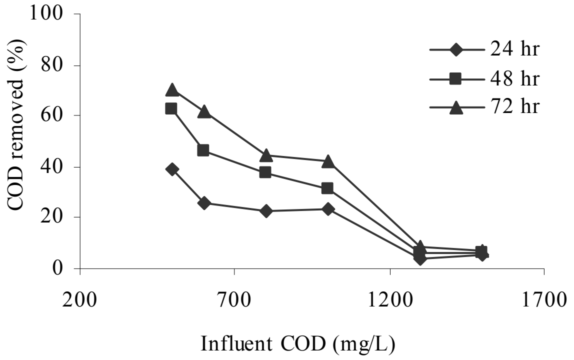

Fig. 5: Variation of organic loading rate and COD removal for pyrogallol wastewater in different detention times

For reactor containing pyrogallol, as shown in Fig. 5, the COD removal efficiency exceeded 80% for influent COD of 600 mg/L after 72 hours. Beyond this there was a steep decrease in COD removal efficiencies. The effect of inhibitory of pyrogallol appeared in influent COD concentrations more than 600 mg/L and with increasing COD loading, longer detention times did not have a remarkable effect to improve this behavior.

*Comparing COD removal efficiency between three phenolic compounds*

From Fig. 6, it can be seen that the COD removal of wastewater containing hydroquinone had better efficiency. The removal efficiency of phenol wastewater with about 20% difference lies lower than hydroquinone. This difference was more apparent in COD ranging between 1000 to 2000 mg/L and reduced by increasing influent COD. As shown in Fig. 5 there is a sudden reduction in COD removal efficiency of wastewater containing pyrogallol comparing to phenol and pyrogallol beyond loading of 600 mg/L. Hence, increasing of influent COD was stopped. More decrease in COD of 1800 mg/L could be related to low DO in the system.

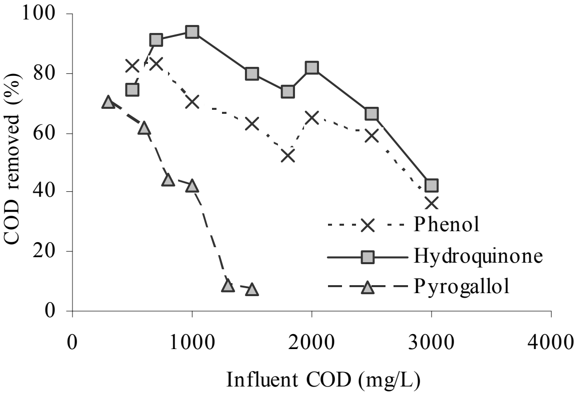

Fig. 6: COD removal efficiency for phenol, hydroquinone and pyrogallol in 72-hr retention time

## DISCUSSION

As mentioned in the introduction part, many researches have been done using chemical treatment for removal of the three compounds used in this study from industrial wastewater. Not much work has been reported for biological removal of these chemicals especially for hydroquinone and pyrogallol. Some of the biological treatment methods which could be comparable with our study are discussed in the following paragraphs:

Two researches are recently done in Sharif University of Technology using MBBR for removal of both molasses and phenol from synthetic wastewater with COD of about 800 mg/L. They have reported 95 percent COD removal efficiency (Hosseini *et al.*, 2001 & 2005). Another study has been shown that after ozonation pretreatment process of the phenol wastewater with COD concentration of 1600 to 6000 mg/L from a petrochemical industry, MBBR with granule activated carbon resulted in 85 to 90 percent removal efficiency (Lin *et al.,* 2001). Not much work has been reported using hydroquinone as a separate wastewater by means of any biological treatment processes whereas it has been identified as a by-product of phenol, nitro phenol and other phenol compounds treatment (Yi *et al.*, 2006).

Biological treatment of pyrogallol in comparison to hydroquinone has not been significantly studied. One of the limited research studies in this area was the use of activated sludge process in treating phenolic wastewater containing pyrogallol. In this study 78 percent of COD and phenols were removed, but not much pyrogallol has been successfully decreased (Pendy *et al.*, 1991). Based on the experimental results of this study, the removal efficiency for each selected compound was affected by the detention time. The three used MBBR systems have shown a proper COD removal efficiency for biodegradation of phenol and hydroquinone wastewater. It has shown that at low phenol concentration (700 to 1000 mg/L) over 80 percent was removed. In addition, by increasing in COD loading rate up to 3000 mg/L, a decrease in COD removal rate was occurred. Similar results (between 92 to 94% at low COD and 80% for COD 1000 to 2500 mg/L) have been observed for hydroquinone wastewater. In the reactor containing pyrogallol (COD 500 mg/L), maximum of 82% removal efficiency was obtained. By increasing COD to 1500 mg/L, because of its toxicity for microorganisms, the removal decreased to 10 percent. In overall, the results have indicated that MBBR has better effect on hydroquinone removal.

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