



Naphthalene degradation in seawater by UV irradiation: The effects of fluence rate, salinity, temperature and initial concentration



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ABSTRACT

A large amount of oil pollution at sea is produced by the operational discharge of oily wastewater. The removal of polycyclic aromatic hydrocarbons (PAHs) from such sources using UV irradiation has become attractive, yet the photolysis mechanism in seawater has remained unclear. This study examines the photodegradation kinetics of naphthalene in natural seawater through a full factorial design of experiments (DOE). The effects of fluence rate, salinity, temperature and initial concentration are investigated. Results show that fluence rate, temperature and the interaction between temperature and initial concentration are the most influential factors. An increase in fluence rate can linearly promote the photodegradation process. Salinity increasingly impedes the removal of naphthalene because of the existence of free-radical scavengers and photon competitors. The results will help understand the photolysis mechanism of PAHs and develop more effective methods for treating oily wastewater generated from offshore industries.

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1. Introduction

The rapid development of ocean industry can affect and alter the marine environment in many ways, of which contamination caused by accidental or operational release of oils draws growing concerns (Li et al., 2012). Such contamination can result in serious, long-term damage to local industries such as fishery and tourism, communities, and the environment (Atlas and Hazen, 2011). Betti et al. (2011) reported that the annual amount of oil entering the global marine environment from sea-based activities was approximately 1.25 million tonnes. While tanker accidents and blowouts at offshore drilling rigs have received the most publicity, they are actually sporadic and only account for a small portion of marine oil pollution (Kadali et al., 2012; Jing et al., 2012a). A disproportionately large amount of oil pollution at sea is produced by the operational discharge of oily wastewater from shipping and offshore oil and gas operations, such as bilge water, ballast water and produced water (Blanchard et al., 2011; Farnen et al., 2010; Jing et al., 2012b). They are usually referred to as “chronic oil pol-

lution” on account of the fact that these discharges occur on a daily basis without triggering a mitigation response. There is a growing recognition that the impacts associated with oily wastewater can be cumulative and longstanding (O'Hara and Morandin, 2010). Owing to the stringent environmental regulations imposed in recent years, a thorough treatment of these waste streams must be carried out prior to discharge.

Oil contained in oily wastewater can be classified into three fractions: dispersed free oil, oil-in-water or water-in-oil emulsions, and dissolved components (Klaassen et al., 2005). While the first two can be mostly removed by gravity separation or hydrocyclone (requires demulsification, if necessary), residual oil droplets and dissolved organic compounds including particularly polycyclic aromatic hydrocarbons (PAHs) would remain unaffected. The occurrence of PAHs is usually of the greatest concern because of their high resistance towards biodegradation, extreme toxicity to marine biota, and possible carcinogenicity and mutagenicity (Tsapakis et al., 2010; Harman et al., 2011). Lee et al. (2011) reported that the accumulation of PAHs can cause severe oxidative DNA damage and therefore leads to tumors and cancer. Further treatment is therefore desirable after oil–water separation to reduce the emission of PAHs.

Removal of PAHs from oily wastewater is possible via many techniques such as biofiltration, biodegradation, adsorption, and phytoremediation (Haritash and Kaushik, 2009). However, most

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of these methods are not directly suitable for marine applications due to space, cost, and safety concerns. UV irradiation has been gaining significant attention and regarded as a promising solution because of its relatively small footprint, low cost and risk, high efficiency as well as its chemical-free nature. In recent years, many previous studies have investigated the degradation of PAHs in water and wastewater by using UV irradiation (Sanches et al., 2011). Woo et al. (2009) characterized the photocatalytic degradation efficiencies and pathways of five major PAHs in aqueous solution. Kwon et al. (2009) studied the degradation of lowly concentrated phenanthrene and pyrene in aspect of kinetics under UV irradiation. The results suggested an inverse relation between the reaction constants and the number of molecules due to agglomeration of hydrophobic molecules in the aqueous environment. Włodarczyk-Makuła (2011) discovered a significant impact on the photolysis of PAHs when changing the exposure time from 10 to 20 s as the removal efficiencies were believed to be proportional to the length and intensity of UV exposure. Salihoglu et al. (2012) applied UV irradiation to remove PAHs from municipal sludge and obtained a removal rate up to 77% after 24 h. The removal rate tended to increase as the temperature elevated during the UV irradiation. However, most of these studies have focused on freshwater systems; the investigation of the photolysis of PAHs in seawater has been limited to a few studies where artificial seawater was mostly used (Kot-Wasik et al., 2004; Saeed et al., 2011; de Bruyn et al., 2012). It is still unclear how efficient UV treatment is in removing PAHs from natural seawater and how changing environmental conditions (e.g., salinity and temperature) can influence the efficiency.

In order to address the research needs and knowledge gaps, this study is to investigate the UV induced degradation kinetics of a typical PAH, namely naphthalene, through a bench-scale reactor. Naphthalene is chosen because it is a major contaminant in oily wastewater such as ballast water, bilge water and produced water, and has been considered as a possible carcinogen to humans. It is relatively more soluble and less hydrophobic than other high molecular weight PAHs which raises a critical bioavailability issue due to its high concentration. Experiments also focus on examining the effects of various factors including UV fluence rate, salinity, temperature and initial concentration.

2. Materials and methods

2.1. Reagents and standards

Naphthalene (>99%) and naphthalene D8 (>99%, internal standard) were purchased from Aldrich and used as received. Dichloromethane (Honeywell Burdick and Jackson, USA) was used for preparing stock solutions and for aqueous sample extraction. Naphthalene spiking solutions were prepared by 1:1 dilution (v/v) of the stock solutions in acetone (Honeywell Burdick and Jackson, USA). The commercial products were used as received without any further purification. Distilled water was produced on-site from a double, fused-silica distillation unit. Natural seawater with a salinity around 25 practical salinity unit (psu) was obtained from a clean coastal site in St. John's, Canada, and the site is considered free of any oil pollution. Seawater was stored and used after filtration (5 μm) to remove suspended solids that could scatter and absorb UV irradiation.

2.2. Photoreactor and light source

The photoreactor used in this study has an inner airtight quartz sleeve and an outer aluminum jacket. The inner sleeve is a clear fused quartz beaker (Technical Glass, USA) with a polycarbonate top lid sealed by a 0.64 cm thick O-ring to avoid the evaporation of naphthalene. A stainless steel stirring rod, attached to a

4.78 cm size stepper motor (4018L-01S-01, from LIN Engineering), is placed at the center of the sleeve. Two stainless steel six-bladed paddle impellers are mounted one above the other on the rod. A 50 W Eheim Jager aquarium heater (3602090, from Eheim, Germany) and a thermometer are deployed to adjust water temperature. Eight 18.4 W low-pressure UV lamps (Atlantic Ultraviolet, Canada) with emission peak at 254 nm were placed around the quartz sleeve (Fig. 1b). The emission peak has a full width half maximum (FWHM) of 15 nm. The outer jacket has an aluminum lid that can be firmly sealed to provide heat and light insulation. Four control buttons are located on the dashboard outside of the jacket body to control lamps in any desired combinations (i.e., two, four, six, and eight lamps).

2.3. UV fluence rate determination

Fluence rate refers to the total radiant power incident from all directions onto an infinitesimally small sphere. According to Bolton and Linden (2003), it is the appropriate term to describe photon intensity in a UV reactor because UV can impinge on the target organic compounds from any direction. For different number of lamps, the fluence rate in the empty quartz sleeve was measured at 17 points on the top, middle, and bottom layers (Fig. 1), respectively, by using a digital UVX Radiometer (UVP, USA). The measurements were made and summed according to the tetrahedral method described by Björn (1995) in order to account for light reflected from all directions. To determine the corresponding fluence rate when the quartz sleeve was filled with sample water, the attenuation coefficient of UV through water (0.0264 cm^{-1}) was taken into account on the basis of Su and Yeh (1995). As most suspended solids were removed prior to the tests, a basic assumption here was that the effective attenuation length was approximated as the internal radius of the reactor (11.1 cm), implying a 25% reduction in all measurements. The adjusted measurements were then interpolated by using a Matlab[®] built-in function (i.e., grid data) on the top, middle, and bottom layers, respectively.

2.4. Experimental procedure

To examine the effects of fluence rate, salinity, temperature, and initial concentration, a full factorial design of experiments (DOE) was employed to determine the significance of each factor being tested as well as their interactions. Two levels of UV fluence rate were obtained by switching on two and six lamps, respectively. Initial naphthalene concentrations were set as 10 and 500 $\mu\text{g L}^{-1}$ which were usually considered as the minimum and maximum values in oily wastewater (OGP, 2002; Johnsen et al., 2004; Blanchard et al., 2011). Temperature was adjusted and maintained at 23 (room temperature) and 40 °C which were reported as the typical temperature range of marine oily wastewater (Levine and Barnes, 2010; Neff et al., 2011; Leichsenring and Lawrence, 2011). Control experiments (without UV irradiation) were conducted at 40 °C to quantify the loss of naphthalene caused by volatilization. Salinity levels of 25, 32.5, and 40 psu were chosen on the basis of the ocean surface salinity at the coastal region of the Grand Bank in Newfoundland (Han et al., 2011) and the mid Atlantic (Qu et al., 2011).

In each of the 24 experimental runs, a volume of 50 μL of naphthalene stock solution (1.2 or 60 mg mL^{-1}) was diluted in acetone (1:1, v/v) and then spiked into 6 L filtered seawater to obtain an initial naphthalene concentration of 10 or 500 $\mu\text{g L}^{-1}$, respectively. Different salinity levels were obtained by evaporating seawater and measured by a salinity meter (Thermo Scientific Orion Star A215). The mixture was vigorously stirred for 20 min to reach the thermal and volatilization equilibria. UV lamps were allowed to warm-up for 20 min before tests. Temperature was controlled by the built-in heater. The paddle impellers were driven by a step-

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