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Study of Kinetics Mechanism of PAHs Photodegradation in Solution

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Abstract

As a typical group of persistent organic pollutants, polycyclic aromatic hydrocarbons (PAHs) are widely distributed and persistent in ecological environment via environmental media (e.g. air, soil, water, suspended solids, organisms, etc.), causing great damage to organisms. Its fate and transportation from surface water to aquifers has attracted more and more attention. As PAHs in surface water or in topsoil will undergo photochemical reactions, a self-designed cylindrical photodegradation reactor has been designed to study the photodegradation of PAHs in aqueous solutions in this study. The results have shown that PAHs degrade rapidly under the lighting conditions. Pseudo-first-order kinetics was observed for degradation of fluorene (FLU), phenanthrene (PHE) and pyrene (PYR), as indicated by linear relations between $\ln(C_0/C_t)$ and time. The reaction kinetics constants of FLU, PHE and PYR were 0.031, 0.045, 0.017 min^{-1} respectively, and the photolysis half-lives of PAHs are only several minutes (FLU, 4.17 min; PHE, 3.79 min; PYR 4.77 min). Their photolysis rates following the decreasing order PHE > FLU > PYR is due to their different absorption spectrum and peak values under the UV-light.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a typical group of persistent organic pollutants (POPs) with two or more fused aromatic rings. As many of them are highly toxic, having carcinogenesis, tetraogenesis and mutagenesis¹, PAHs have received much concern in recent years. PAHs mainly derive from incomplete combustion

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or thermal decomposition of fossil fuels, such as oil, coal, wood, organic polymer compounds, crop straw and other substances containing hydrocarbons². They are ubiquitous in atmospheres, surface water, sediments and particulate matters³. PAHs in surface water or in topsoil will undergo photochemical reactions, including photo-dimerization and photo-oxidation⁴. It is known that photodegradation process plays an important role in the degradation of PAHs in water by highly generating reactive intermediates among which hydroxyl radical is predominant. Besides, some bio-refractory PAHs can be easily degraded by sunlight⁵. To ultimately figure out PAHs' fate and transportation in surface water before they enter into aquifers, we studied the kinetics mechanism of the photodegradation of three kinds of PAHs *fluorene (FLU)*, *phenanthrene (PHE)* and *pyrene (PYR)* in aqueous solutions as a function of solution medium.

2. Materials and Methods

2.1. Experimental apparatus and reagents

A self-designed cylindrical photodegradation reactor was used (Fig. 1); simulated lighting was performed by a 10 W low-pressure UV mercury vapor lamp (peak emission wavelength 254 nm, outer diameter 2.5 cm, length 35 cm); fluorene (FLU), phenanthrene (PHE) and pyrene (PYR) were purchased from AccuStandara (USA); organic solvents including dichloromethane (DCM) and n-hexane (analytical grade) were purchased from CNWBOND (Germany); anhydrous sodium sulfate was heated in a muffle furnace at 450 °C for 4 h and stored in a sealed desiccator prior, etc. All glassware was cleaned with sulfuric acid potassium dichromate solution, washed with water, and dried at 180 °C for 4 h.

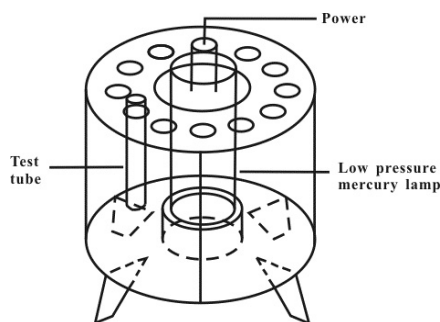


Fig.1. diagram of the self-designed photodegradation reactor

2.2. Photodegradation experiments

The photodegradation experiments were carried out using a self-designed reactor in darkness. 10 mL aqueous solution with PAHs (2 mg/L FLU, 1 mg/L PHE, 0.14 mg/L PYR) was transmitted to the quartz tubes (approximate volume 20 ml). Quartz is a preferable material for photo-irradiation as it is transparent to the solar radiation (UV-A) reaching the earth surface. The reaction tubes were placed in the self-designed reactor. A 10 W low-pressure UV mercury vapor lamp was selected to provide a source of light. After specified time (0, 5, 10, 20, 30, 60 and 120 min), the irradiated quartz tube containing PAHs sample was removed to process for PAHs determination.

2.3. PAHs analysis

PAHs concentration was determined by means of an Agilent GC 6850 gas chromatograph equipped with a DB-5 MS capillary column (30 m × 250 μm film thickness × 0.25 mm, Agilent), operating with helium carrier gas, coupled to an Agilent MD 5975 mass spectrometer (MS). The GC injector was operated in splitless mode, 1 μL aliquots were injected using an autosampler. GC oven was programmed to hold 80 °C for 1 min, then the temperature was elevated by 10 °C min⁻¹ to 200 °C, which was held for 2 min. The temperature was finally brought

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