601d Ultrasonic Degradation of Phenol in the Presence of Composite Particles of Tio_2 and Activated Carbon

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Introduction

Titania (TiO_2) has recently used as a heterogeneous catalysis for the degradation of organic compounds with light irradiation. When energy is supplied to the TiO_2 particles, the positive hole generated in particles reacts with the water in the vicinity of the particle surface to form OH radical, which is thought to promote the degradation reaction. The UV light has been normally used as an energy source. However, the light is screened by particles themselves and the region exhibiting the catalytic property is spatially limited in the reactor, so that the particle concentration cannot be raised. An ultrasonic irradiation might be used as an alternative energy source for formation of OH radical. The ultrasonic wave propagates throughout a reactor, so that it is possible to increase the catalyst particle concentration.

Most of organic compounds are hydrophobic and are hard to close to the hydrophilic TiO_2 surface and exist in bulk water phase. If TiO_2 is immobilized on the activated carbon, which is generally used for the adsorption of organic compounds, the distance between organic compounds and OH radicals is short. OH radicals are effectively used for the degradation and the rate of degradation of organic compounds is expected to be high.

In this research, the ultrasonic degradation of phenol in water was carried out in the presence of the composite particles of anatase-type TiO_2 and activated carbon. The effects of the added amount of the particles and the AC/TiO_2 ratio in the composite particles were also investigated.

Preparation of composite particles and degradation experiment

The composite particles of TiO_2 and activated carbon were prepared by using sol-gel method. Titanium tetraisopropoxide was hydrolyzed to form titanium dioxide gel. Activated carbon and anatase-type TiO_2 particles were added to the gel solution to immobilized TiO_2 onto the activated carbon using TiO_2 gel as a binder. The prepared particles were dried at 423 K. In 25 cm^3 of 1 mol/m^3 phenol aqueous solution, a specified amount of the particles were suspended. The degradation of phenol was carried out at 293K with irradiating the ultrasound at a frequency of 20 kHz and an output power of 75 W without light irradiation. The concentration of phenol in the water phase was directly measured by HPLC. In the particle suspension system, phenol exists both water and particle phases. The amount of the adsorbed phenol on the particle phase was estimated using adsorption equilibrium relation of a Langmuir-type equation between water and particle phases. The residual amount of phenol in the reactor was the summation of those in water and particle phases.

Results and Discussion

Figure 1 shows the time course of the phenol concentration in the water phase from the beginning of the adsorption. After 180 minutes had passed, the ultrasonic irradiation was started. Ultrasonic irradiation time is indicated in parentheses. The phenol concentration monotonically decreased for all conditions. In cases that TiO_2 particles were suspended, the phenol concentrations were almost same as those without TiO_2. The lower phenol concentration in the presence of the activated carbon or composite particles was responsible for the adsorption of the phenol on the activated carbon. However, it is difficult to compare the results for the composite particles with that for activated carbon only. To evaluate the true residual concentration of phenol, i.e., the phenol concentration in the system, it is necessary to estimate the amount of phenol adsorbed on the particles as well as that in the liquid phase.

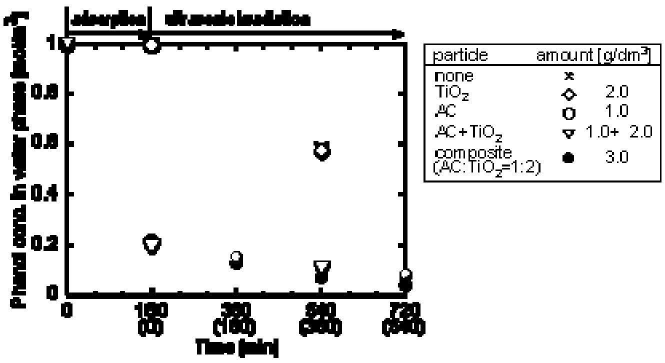

Fig.1 Time course of the phenol concentration in liquid phase.

A Langmuir isotherm equation was given as Eq.(1).

$$q = \frac{q_0 KC}{1+KC} \quad (1)$$

Figure 2 shows the Langmuir plots for the activated carbon and the composite particles. The adsorption equilibrium constant *K* and maximum adsorption capacity q_0 estimated from the linear regression analysis were shown in Table 1.

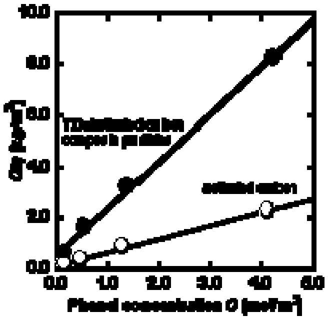

Fig.2 Langmuir plot for activated carbon and composite particles.

Table I Estimated constants.

particle	K [m^3/mol]	q_0 [mol/kg]
activated carbon	3.16	1.88
composite	3.05	0.59

By using the adsorption equilibrium equation, the amount of phenol adsorbed on the particle was estimated from the phenol concentration in the liquid phase. Then, the phenol concentration in the system, the sum of the phenol concentration in the water phase and that on the solid phase, was calculated.

Figure 3 shows the time course of the phenol concentration in the system from the beginning of the ultrasonic irradiation. In the presence of either activated carbon or TiO_2, the phenol concentrations were almost same as those without TiO_2. In case that both TiO_2 and activated carbon were simply suspended, the phenol concentrations were also the same as those without TiO_2. On the other hand, the phenol concentrations for the composite particles were much lower than those for the activated carbon. This is because the distance between phenol on the activated carbon and OH radical generated by TiO_2 is short and it is easy for the radical to react with phenol. These results suggest that the ultrasonic irradiation to the composite particles was highly effective for the degradation of phenol.

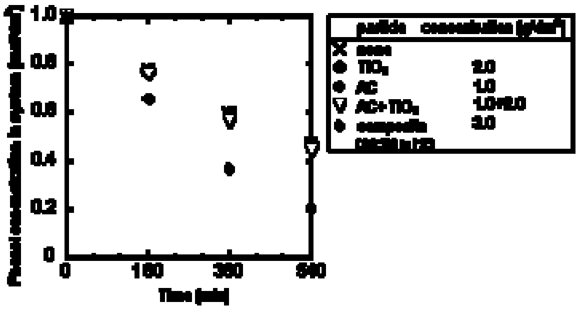

Fig.3 Time course of phenol concentration in system.

Figure 4 shows the effect of added amount of the particles on the time course of the phenol concentration in the system. When the amount of the composite particles was higher, the phenol concentration was lower. At 90 g/dm^3 of composite particles, phenol was disappeared within 3 hours. It is concluded that the higher concentration of composite particle addition with ultrasonic irradiation are very effective for enhancement of the degradation of phenol.

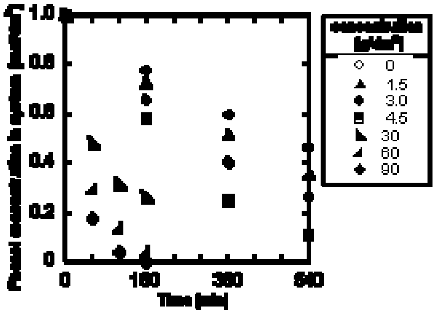

Fig.4 Effect of particle concentration on time course of phenol concentration in system.

Figure 5 shows the effect of AC/TiO_2 ratio of the composite particles on the time course of the phenol concentration in the system. Higher TiO_2 ratio of the composite particles results in lower phenol concentration. The amount of generated OH radical per particles is larger for higher TiO_2 ratio.

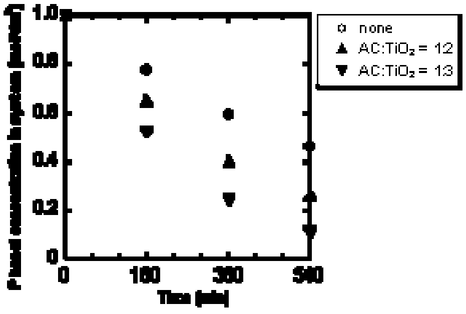

Fig.5 Effect of AC/TiO_2 ratio in the composite particles on time course of phenol concentration in system.