

# Atrazine adsorption from aqueous solution using powdered activated carbon—Improved mass transfer by air bubbling agitation

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## Abstract

A set of batch adsorption kinetic tests of atrazine adsorption by powdered activated carbon (PAC) was performed using air bubbling as the mixing method. It was found that air bubbling at appropriate rates could achieve good mixing. Even a slight turbulence generated by a few bubbles could provide a reasonable mixing to facilitate PAC adsorption compared with unstirred processes.

The estimated mass transfer coefficient in the liquid film surrounding the PAC particles increased linearly with the increase in air bubbling rate up to a plateau value. The experimental mass transfer coefficients in the bubbling system compared favorably with values calculated using correlations developed for conventional magnetic stirring systems, with the help of a conversion of the bubbling rates to the equivalent stirring speeds.

The effect of intermittent air bubbling on the adsorption rate was also tested by generating bubbles intermittently at different net air flow rates. It was found that at the same net flow rate, intermittent higher intensity sparging could be more efficient for the PAC adsorption than continuous lower intensity sparging. This suggests that intermittent high intensity bubbling is the preferable operation, with the potential not only to assure good PAC adsorption efficiency but also to reduce the air/energy consumption.

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**Keywords:** Air bubbling; Powdered activated carbon; Mass transfer coefficient; Intermittent bubbling

## 1. Introduction

For trace organics removal from aqueous solution, the combination of powder activated carbon (PAC) adsorption with micro-filtration (MF)/ultrafiltration (UF) membranes is attractive as it takes advantages of both processes to treat the organics [1–3]. The organic solutes can be adsorbed onto the PAC particles with high efficiency, while the fine PAC particles can be separated from the treated water by membrane filtration [4,5]. Submerged membranes can be used for the combined process with PAC in suspension. In this case, the membrane is mounted directly in the adsorption vessel, and air bubbling can be injected into the vessel to provide mixing for adsorption and to improve the filtration efficiency by creating turbulence for control of membrane fouling [6]. Moving bubbles generate shear and liquid movement that can disrupt the concentration polarization layer or cake for-

mation, diminish the boundary layer resistance and accordingly, increase the permeability [7] and prolong the operating lifespan [8].

Although the effect of bubbling on membrane processes has been actively studied and reviewed [6], the role of bubbling on the PAC adsorption process is not yet elucidated. The adsorption process consists of four consecutive steps [9]: (1) external diffusion in the bulk solution; (2) diffusion in the liquid film surrounding the carbon particle; (3) surface diffusion through the pores of the carbon; and (4) the contaminant being adsorbed onto the active sites in the micropores. Of these, bubbling may have an effect on the first two steps.

Mass transfer in gas–liquid–solid systems has been intensively studied in chemical engineering for decades [10–13]. However, different from a typical three-phase system, the bubbling applied to the PAC-submerged membrane system acts only to promote mixing and turbulence without involving mass transfer in the gas phase. The situation is similar to a previous study [14] on the rate of solid–liquid mass transfer with interfacial bubble generation, where mass transfer took place only at the

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### Nomenclature

$a$	specific interfacial area based on the liquid volume ( $\text{m}^{-1}$ )
$C_0$	initial adsorbate concentration in bulk solution ( $\mu\text{g/L}$ )
$C_t$	adsorbate concentration in bulk solution at any time $t$ ( $\mu\text{g/L}$ )
$C_{st}$	liquid phase adsorbate concentration at solid–liquid interface ( $\mu\text{g/L}$ )
$d_a$	stir bar diameter (m)
$d_p$	carbon particle diameter (m)
$D_v$	volumetric diffusivity for dilute liquid solutions ( $\text{cm}^2/\text{s}$ )
$K_1$	mass transfer coefficient in liquid film ( $\text{cm}/\text{min}$ )
$M_B$	molecular weight of solvent ( $\text{g}/\text{mol}$ )
$n$	stirring speed (rpm)
$Q_b$	bubbling rate ( $\text{L}/\text{min}$ )
$Re$	Reynolds number
$Sc$	Schmidt number
$Sh$	Sherwood number
$T$	absolute temperature (K)
$t$	operation time (min)
$u_0$	velocity of approaching stream ( $\text{m}/\text{s}$ )
$u_m$	the magnetic stirring speed (rpm)
$V_A$	molar volume of solute as liquid at its normal boiling point ( $\text{cm}^3/\text{gmol}$ )

### Greek letters

$\mu$	viscosity of solution ( $c_p$ , $10^{-3} \text{ kg/ms}$ )
$\rho$	density of solution ( $\text{kg}/\text{m}^3$ )
$\psi_B$	association parameter for solvent, for water $\psi_B = 2.6$

solid–liquid interface and no mass was exchanged with the bubbles. This study referred to: (a) the ascending bubble swarm causing an upward flow which reduces the diffusion layer thickness at the solid surface, and (b) the detached bubbles inducing radial momentum transfer which brings a fresh supply of the liquid reactant to the solid surface. Although their discussions were based on a larger flat solid surface, it may still assist on understanding the effect of upward bubble motion on the mass transfer in the suspended PAC particle–liquid system. Other researchers [15] have reported that the liquid–solid mass transfer coefficients increased with gas velocity up to  $6 \text{ cm/s}$ , but were constant at higher gas throughputs. The particle Reynolds number ( $Re$ ) was correlated to an energy dissipation rate to avoid the difficulty of estimating the relative velocity between the suspended particle and the fluid. This method is more suitable for relatively high Reynolds numbers ( $>1500$ ), which are not appropriate to our low air flows and small particle sizes. However, we have observed improved adsorption kinetics by bubbling in our previous studies [16], provided no other impurity was introduced during the process of air bubbling.

The enhancement of liquid–solid mass transfer by mechanical stirring has already been well studied [17]. Since the air bubbling in our system plays a similar role to mechanical stirring in enhancing mixing, we aim to link the superficial gas bubbling velocity to a mechanical stirring speed by a mixing experiment. Assuming a relationship between mixing and liquid flow patterns and mass transfer we then use mass transfer correlations developed for conventional mechanically stirred systems to determine the effect of bubbling on mass transfer from liquid to PAC particles. In addition, as intermittent bubbling can be used to improve membrane performance instead of continuous bubbling to reduce energy consumption and cost, the effect of this approach on the PAC adsorption kinetics has been examined. This study should provide insight into the preferred bubbling conditions that could be applied in the hybrid membrane adsorption system process.

## 2. Background

The dynamics of the adsorption of organic compounds onto activated carbon in water can be described using the well-developed homogeneous surface diffusion model (HSDM) [18]. The model incorporates the liquid film mass transfer coefficient and the surface diffusion coefficient to describe the overall carbon adsorption process. It is commonly recognized [9] that the adsorption rate is controlled by boundary layer film diffusion in the initial stages of adsorption in batch reactors. Eventually, as the carbon becomes loaded with the adsorbates, surface diffusion becomes the controlling factor. Therefore, to observe the bubbling effect on the mass transfer in the liquid film, an alternative to fitting data to the complex HSDM is to use a simple approach based on the general mass balance in a batch kinetic adsorption test to describe only the initial stage of an adsorption process. For the simplified approach we assume that film diffusion is the rate-controlling step in the initial stage of atrazine adsorption [19], and thus, the rate of concentration change is approximately equal to the film diffusion rate. The atrazine concentration change in the bulk liquid with adsorption time can then be described using the following equation:

$$\frac{dC_t}{dt} = -K_1 a (C_t - C_{st}) \quad (1)$$

where  $C_t$  is the adsorbate concentration in bulk solution at time  $t$ ,  $C_{st}$  the liquid phase adsorbate concentration at the solid–liquid interface,  $K_1$  the mass transfer coefficient in the liquid film and  $a$  is the specific interfacial area based on the liquid volume for a completely mixed batch reactor. At the beginning of the kinetic test,  $C_{st}$  is near zero and Eq. (1) can be simplified as:

$$\frac{dC_t}{dt} = -K_1 a C_t \quad (2)$$

Solving the first order Eq. (2), the  $K_1$  can be written as:

$$K_1 = -\frac{1}{at} \ln \left( \frac{C_t}{C_0} \right) \quad (3)$$

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