

《原著》

## 흡착제 크기에 따른 GAC의 멜라민 흡착 처리 : 반응속도와 분산-확산

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### Treatment of Melamine by GAC Adsorption According to Adsorbent Size: Kinetics and Dispersion-Diffusion

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#### ABSTRACT

Adsorption of melamine was examined using columns packed with granular activated carbon (GAC). Raw GAC was sieved with 20, 40, 60 and 80 mesh to determine the influence of adsorbent particle size on reaction and diffusion. The mass ratio of the adsorption capacity of GAC for melamine ranged from 9.19 to 11.06%, and adsorption rates increased with decreasing particle size within this range. Rate constants between 3.295 ~ 4.799 min<sup>-1</sup> were obtained using a pseudo-first-order equation that was used to determine adsorption kinetics. A surface diffusion model was adapted to take into account the unsteady-state equation of a spherical adsorbent by converting the surface concentration from a constant to a variable governed by a dispersion equation. The calculated values were fit with the experimental results by using the diffusion coefficients as regression parameters. The modified equation exhibited a more precise agreement with respect to the sum of the absolute error (SAE).

**Key words :** Melamine, Adsorption, GAC, Surface diffusion, Crank's diffusion equation

#### 1. Introduction

The China milk scandal of 2008, in which milk products were tainted with melamine, generated a significant amount of worry and nervousness around the globe (Xiu and Klein, 2010; Pei et al., 2011). Consumption of melamine can cause kidney stones, and acute melamine toxicity can be fatal (Dobson et al., 2008; Puschner et al., 2007).

However, owing to its excellent fire retardant properties, melamine is produced industrially in significant quantities. Thus, there is an ongoing possibility of contamination from the effluent of melamine-producing facilities (Lee et al., 2011).

Ground water and surface water are closely related with various factors (Lee, 2004), so the contamination of surface

water could affect ground water directly or indirectly. Actually, there have been several reports on the melamine contamination of groundwater near chemical plants (Malina, 2004; Alink et al., 2007).

Despite these concerns, there have been few studies concerning the treatment of melamine to mitigate possible contamination. Xu et al. (2013) tracked the fate of melamine in an activated sludge process, and they concluded that melamine does not adequately biodegrade. Although the study included a long-term sludge adaptation, melamine inhibited self-degradation in a Modified Ludzack-Ettinger (MLE) process and continuous stirred tank reactor (CSTR) process. Even when injected in the middle of the treatment process, melamine showed poor

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biodegradability.

However, another study showed that melamine was strongly adsorbed onto carbon particles. From the measured results of the bonding energy by local density approximation, aminotriazines showed strong interactions with carbon particles with an increased number of  $\text{NH}_2$  groups added (Wuest and Rochefort, 2010). Melamine is an aminotriazine with three  $\text{NH}_2$  groups. Thus, activated carbon adsorption seems to be suitable for the treatment of melamine. In this study, a column reactor of granular activated carbon (GAC) was adopted to utilize the adsorption of melamine to a carbon adsorbent.

We recently reported on the adsorption of melamine to GAC (Lee et al., 2011), demonstrating the effectiveness of the removal of melamine using GAC. The model used to interpret the adsorption was a deactivation factor.

The treatment of GAC considering adsorbent size is usually explained by adsorption models and agrees well with experimental results (McKay et al., 1979; Schouten et al., 2009). There are two main approaches to the adsorption model of GAC using the size of the adsorbent: the kinetic model, such as the pseudo-n-th order model and Weber Morris plot (Wasewar et al., 2009), and the white model, which is a physical and mechanical deviation (Sontheimer et al., 1988; Yoon, 1999). The white model of adsorption is a combination of the partial differential equations for dispersion and diffusion. To solve these equations, the finite element method or finite difference method is widely used. The diffusion equation was solved algebraically by Crank with regard to one adsorbent particle (Crank, 1965); however, in a column reactor, a dispersion equation is also needed.

Thus, in this study, GAC adsorption was evaluated as a potential treatment for melamine. Specifically, samples were passed through columns packed with GAC particles sieved with 20, 40, 60 or 80 mesh, and the effluent was examined to determine the effect of adsorbent size on adsorption. The adsorption rate according to the size of the adsorbent was interpreted with pseudo-first-order kinetics and the dispersion-diffusion model. In this process, Crank's algebraic solution of the diffusion equation was used and modified with the dispersion equation using the finite difference method.

## 2. Materials and methods

### 2.1. Feeding solution

A feeding solution of melamine (200 mg/L) was prepared using a standard melamine reagent (> 99% purity) purchased from Sigma-Aldrich.

### 2.2. GAC columns

Columns were packed with a GAC adsorbent (Norit 1240, Norit, Netherlands) sieved with 20, 40, 60 or 80 mesh. The properties of GAC showed a negligible difference according to pore size, which was thus expected to have no significant effect. The average values of the specific surface area, pore volume and average pore size for the sieved GAC were  $1088 \text{ m}^2/\text{g}$ ,  $0.63 \text{ (cm}^3/\text{g at } p/p_0=0.99)$  and  $4.2 \text{ nm}$ , respectively.

For each size, 3 g of Norit 1240 was packed into a column with a capacity of 1.56 ml. The feeding solution was passed through columns at a flow rate of 3.5 ml/min. Effluents were obtained every 20 minutes except for the first two intervals, which were collected every 10 minutes.

### 2.3. Analysis

First, the samples of the effluents were prepared in a solid phase extraction (SPE) clean-up process. The obtained samples were activated and washed with acetonitrile (ACN) in Oasis MCX 6cc 150 mL cartridges. ACN was obtained from Samchun (HPLC grade, Korea). The eluant of 160  $\mu\text{L}$  from the cartridge placed in a gas chromatograph (GC) vial was vaporized using  $\text{N}_2$  gas. To the vial, 200  $\mu\text{L}$  of N,O-Bis trifluoroacetamide (BSTFA) with 1% trimethylchlorosilane (TMCS) and 200 chlorointernal standard were added, stirred vigorously and kept at  $70^\circ\text{C}$  for 45 minutes. BSTFA with 1% TMCS was purchased from Tokyo Chemical Industry (derivatization grade, Japan).

The samples were analyzed using a 6890N GC with a Pegasus HT time of flight (TOF) detector. For analysis, we used an Agilent DB-5MS column ( $30 \text{ m} \times 0.25 \text{ nm} \times 0.25 \text{ um}$ ) with helium gas at a flow rate of 1.2 mL/min. The injection temperature and pressure were  $280^\circ\text{C}$  and 12.9 psi, respectively. The temperature of the transfer line was set at  $290^\circ\text{C}$ . The oven program was set to hold  $100^\circ\text{C}$  for 1 minute and increase at  $10^\circ\text{C}/\text{min}$  to  $210^\circ\text{C}$ . The times for

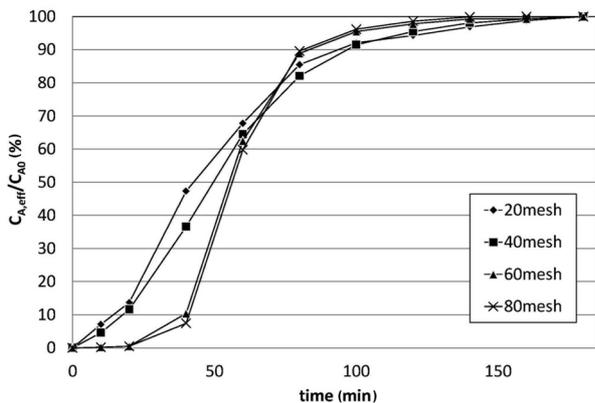


Fig. 1. Melamine effluent fractions from various sized adsorbents as a function of operating time.

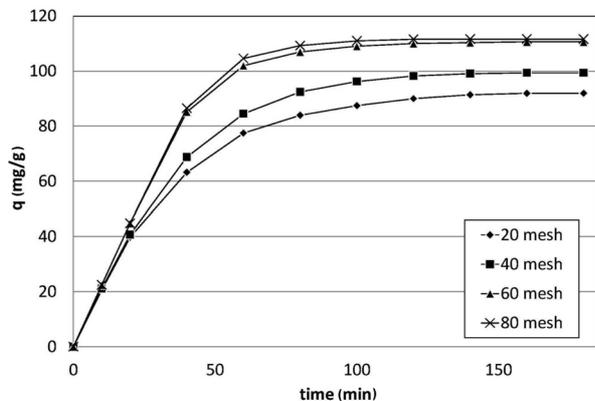


Fig. 2. Total transfer of melamine to GAC of various sized adsorbents as a function of operating time.

detection and retention were 12 and 11.5 minutes, respectively.

### 3. Results

#### 3.1. Effluent fraction and total transfer to GAC

The effluent fraction of melamine and the total transfer to GAC with respect to operating time are shown in Fig. 1 and Fig. 2, respectively.

Smaller sized adsorbents exhibited a more effective performance. To confirm this relationship in detail, we applied kinetics and diffusion models.

The adsorption capacities of GAC for melamine in terms of mass ratio were 9.19, 9.94, 11.06 and 11.16% for the 20, 40, 60 and 80 mesh, respectively. The mass ratio is the total amount of adsorbate relative to the adsorbent with a unit of weight percent. In general, diffusion increased with

Table 1. Variation of the rate constant  $k_1$  and RSQ as a function of adsorbent size for linear regression

	$k_1$	RSQ
20 mesh	0.03295	0.9698
40 mesh	0.03695	0.9856
60 mesh	0.04220	0.9982
80 mesh	0.04799	0.9618

decreasing adsorbent size, while adsorption capacity increased.

#### 3.2. Kinetics Model: Pseudo-first-order

To examine the adsorption with respect to transfer onto GAC, we adapted a pseudo-first-order equation as the kinetics model (Adriano et al., 2005).

$$\frac{dq}{dt} = k_1 (q_e - q) \quad (1)$$

Applying initial conditions of  $t = 0, q = 0$  and  $t = t, q = q$ , Equation 1 generates a regression line after definite integration.

$$\log(q_e - q) = \log q_e - \frac{k_1}{2.303} t \quad (2)$$

Experimental results were adapted using Equation 2 (Table 1). The reaction rate constant increased as the size of the adsorbent decreased, as shown in Table 1.

##### 3.2.1. Effect of adsorbent size

A plot of  $\log k_1$  vs.  $\log r_s$  is shown in Fig. 3. The mean particle diameter of the adsorbent size range,  $r_s$ , was determined using a representative value for each mesh number. Linear regression of the data in Fig. 3 produced a high RSQ value of 0.9678, indicating a strong relationship between adsorbent size and reaction constant.

#### 3.3. Dispersion and Diffusion Models

##### 3.3.1. Approximation of Crank's equation

In the present study, we used a surface diffusion model (Sontheimer et al., 1988) which presumes that the adsorbent has a homogeneous spherical structure. The diffusion model is described in Equation 3.

$$\frac{\partial q_A}{\partial t} = D \left( \frac{\partial^2 q_A}{\partial r^2} + \frac{2}{r} \frac{\partial q_A}{\partial r} \right) \quad (3)$$

Crank developed an analytical solution to Equation 3

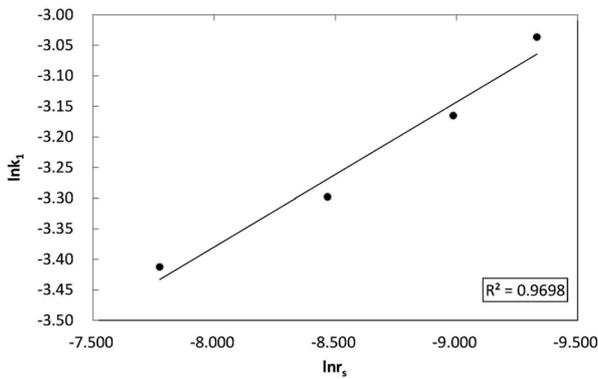


Fig. 3. Log rate constant ( $k_1$ ) vs. log particle radius ( $r_s$ ).

(Crank, 1965) as follows:

$$q_A = q_A^* + \frac{2rs}{\pi} (q_{A0} - q_A^*) \sum_{n=1}^{\infty} \frac{(-1)^{n+1}}{n} \frac{1}{r} \sin\left(\frac{n\pi r}{r_s}\right) \exp\left(\frac{-Dn^2 \pi^2 r}{r_s^2}\right) \quad (4)$$

where  $q_A^*$  is the surface concentration at equilibrium. In a batch system, the surface concentration can be determined using the influent concentration. Crank derived this model by holding  $q_A^*$  constant. However, in the present study, the adsorbent was packed in the shape of the column, and as a result, the adsorbate approaching at the surface was influenced by a dispersion mechanism. Therefore, the surface concentration  $q_A^*$  was not constant and varied with time as follows:

$$q_{A,t}^* = q_{A,t}^*(t) \quad (5)$$

As a result, the analytical form of Equation 4 could not be used in our study. Specifically, Equation 3 incorporating Equation 5 could not be solved analytically as in Equation 4. Thus, several assumptions were made in order to obtain an analytical solution as in Equation 4, including an adsorbed concentration that decreased exponentially with time. Therefore, the influence of the surface concentration,  $q_A^*$ , decreased with time. Thus, all of the terms in Equation 4, except for  $q_A^*$ , decreased exponentially. By applying this approximation and including the term  $q_{A,t}^*$ , the average concentration throughout the spheres at time  $t$  is as follows (Appendix 1).

$$\dot{q}_A = q_{A0} - \frac{3}{4\pi r_s^3} \cdot 8\pi r_s D \int_0^t (q_{A0} - q_{A,t}^*) \sum_{n=1}^{\infty} \exp\left(\frac{-Dn^2 \pi^2 r}{r_s^2}\right) dt \quad (6)$$

Table 2. Dispersion coefficients and film mass coefficients as a function of adsorbent radius

	$D_L$ (m <sup>2</sup> /s)	$k_f$ (m/s)
20 mesh	$1.631 \times 10^{-8}$	$6.384 \times 10^{-8}$
40 mesh	$1.604 \times 10^{-8}$	
60 mesh	$1.591 \times 10^{-8}$	
80 mesh	$9.569 \times 10^{-9}$	

Equation 6 represents the final modified form. The methods used to estimate  $q_{A,t}^*$  are described below.

### 3.3.2. Dispersion Model

We presumed that the arrival of the adsorbate to the external surface of the adsorbent was governed by the following dispersion equation (Sontheimer et al., 1988).

$$\frac{\partial C}{\partial t} = D_L \frac{\partial^2 C}{\partial z^2} - u \frac{\partial C}{\partial t} - \left(\frac{1 - \varepsilon_b}{\varepsilon_b}\right) \left[ \frac{3k_f}{r_s} (C(z,t) - C_{r=r_s}) \right] \quad (7)$$

To calculate the surface concentration, an infinite difference method was used, whereby the initial condition was set as follows:

$$C(t, z=0) = C_0 \quad (8)$$

Most importantly, the adsorbate concentration of the external surface,  $C_{r=r_s}$ , could be obtained by the following boundary condition deviated from the surface diffusion model (Appendix 2) using the infinite difference method.

$$\frac{\partial C_r}{\partial t} = \frac{2}{r} k_f (C - C_{r=r_s}) \quad (9)$$

Thus, the average adsorbate concentration of the bulk phase along the longitude axis was obtained using an infinite difference method and curve fitting with the effluent concentration. The parameters  $D_L$  and  $k_f$  are shown in Table 2. The film mass coefficient,  $k_f$ , was fixed for all cases.

The value of  $D_L$  increased with increasing adsorbent particle size, and thus it appeared that larger particle sizes led to more spacious pores.

The adsorbate concentration of the external surface was converted to the amount adsorbed onto the adsorbent using an isotherm equation. Specifically, the relationship between the concentration outside of the spheres and the adsorbed concentration within the spheres was obtained by the

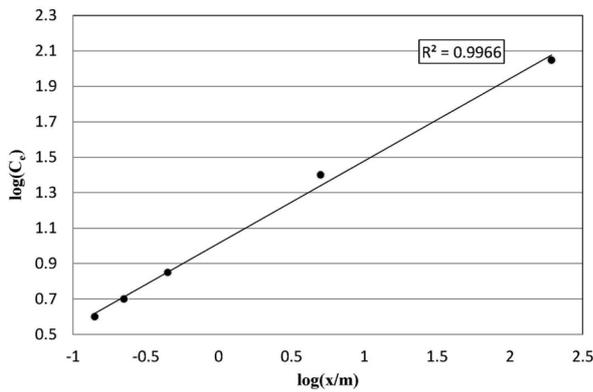


Fig. 4. Freundlich isotherm curves for adsorption of melamine by GAC.

following Freundlich isotherm:

$$q = kC_r^{1/n} \tag{15}$$

The results of the Freundlich isotherm for melamine adsorption on GAC are shown in Fig. 4.

Based on the results of Fig. 4, the Freundlich parameters  $k$  and  $1/n$  were determined as 10.327 and 0.465, respectively. Next, an estimated value for  $q$  from Equation 6 was obtained by substituting the calculated value using the infinite difference method and isotherm for  $q_t^*$ . Numerical integration was then used to evaluate Equation 6.

Table 3. Diffusion coefficients and SAE for varying particle sizes

Mesh	Modified equation		Original equation	
	$D$ m <sup>2</sup> /s	SAE	$D$ m <sup>2</sup> /s	SAE
20	$3.591 \times 10^{-12}$	41.400	$2.586 \times 10^{-12}$	49.193
40	$1.390 \times 10^{-12}$	38.674	$8.502 \times 10^{-13}$	51.808
60	$1.080 \times 10^{-12}$	40.988	$5.983 \times 10^{-13}$	54.220
80	$5.645 \times 10^{-13}$	42.617	$3.230 \times 10^{-13}$	47.850

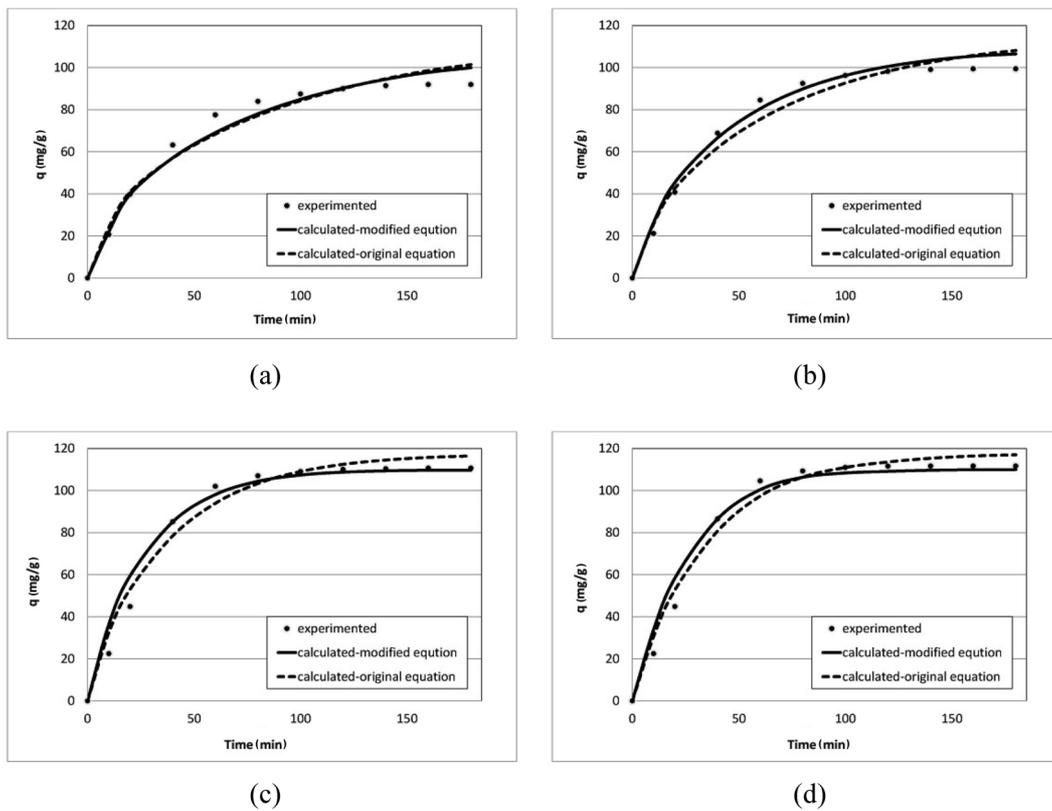


Fig. 5. Comparison of experimental data with the amount of adsorbate adsorbed according to the modified and original equations: (a) 20 mesh, (b) 40 mesh, (c) 60 mesh, (d) 80 mesh.

### 3.3.3. Diffusion coefficient

Regression curve fitting and error analysis were used for the experimental data and the results calculated from Equation 6 to obtain best-fit diffusion coefficients. The sum of the absolute error (SAE) was used for error analysis (Wasewar et al., 2009) as follows:

$$SAE = \sum_{i=1}^n |q_{estm} - q_{exp}|_i \quad (16)$$

Diffusion coefficients from Equation 4 were also obtained to compare the level of agreement between values calculated with the modified equation and original equation, Equation 17 (Crank, 1965). The modified equation gave more precise results with respect to SAE (Table 3).

$$\dot{q}_A = q_{A0} - (q_{A0} - q_{A}^*) \left[ 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{\pi^2} \exp\left(\frac{-Dn^2 \pi^2 r}{r_s^2}\right) \right] \quad (17)$$

We expected that smaller sized adsorbents would have higher diffusion coefficients because of their larger reaction rate constant. However, the diffusion coefficient  $D$ , shown in in Table 3, did not increase with decreasing adsorbent radius. This finding was attributed to the sphere used in the study not being an independent adsorbent, but rather a small particle packed in a column as part of the adsorption as a whole.

The total number of particles that can be placed in a column increases as particle radius decreases. Thus, the smaller sized adsorbents, which had low diffusion coefficient values, exhibited higher reaction rate constants. Fig. 5 shows the comparison between experimental data and calculated results for each case.

## 4. Conclusions

Melamine adsorption treatment was performed using a GAC adsorbent. To examine the effect of adsorbent size on melamine adsorption, we used GAC sieved with 20, 40, 60 or 80 mesh. We observed that the adsorption rate increased as the size of the adsorbent decreased.

The adsorption phenomenon was interpreted using the dispersion equation and diffusion model described by Crank. We assumed that the surface concentration in the Crank equation changed according to time, and the model

was modified accordingly. Experimental results were fit to the calculated data using diffusion coefficients as regression parameters. The values calculated from the modified equation were compared with those generated using Crank's original equation. The results of the modified equation were more precise than those determined by SAE.

### Notations

$C$ : Adsorbate concentration, mg/L

$C_{A, eff}$ : Effluent concentration, mg/L

$C_{A0}$ : Influent concentration, mg/L

$C_e$ : Equilibrium concentration, mg/L

$C_r$ : Adsorbate concentration of the external surface of an adsorbent, mg/L

$D$ : Diffusion coefficient,  $m^2/s$

$D_L$ : Dispersion coefficient,  $m^2/s$

$k_j$ : Rate constant of the pseudo-first-order model,  $min^{-1}$

$k_f$ : Film mass transfer coefficient, m/s

$q$ : Amount of adsorbate adsorbed at time t, mg/g

$q_e$ : Equilibrium adsorption capacity, mg/g

$q_A$ : Concentration inside the spheres, mg/g

$q_{A0}$ : Initial concentration inside the spheres, mg/g

$q_{A}^*$ : Surface concentration of spheres as a constant, mg/g

$q_{A,t}^*$ : Surface concentration of spheres as a function of time, mg/g

$\bar{q}_A$ : Average concentration throughout the spheres at time t, mg/g

$r$ : Sphere distance, m

$r_s$ : Adsorbent radius, m

$t$ : Time, min

$x/m$ : Amount of adsorbate per unit of adsorbent, mg/g

$\varepsilon_b$ : Porosity of a packed bed

### Appendix 1

Crank's solution was transformed as follows by converting the surface concentration from a constant to a variable from Equation 5.

$$q_A \approx q_{A,t}^* + \frac{2r_s}{\pi} (q_{A0} - q_{A,t}^*) \sum_{n=1}^{\infty} \frac{(-1)^{n+1}}{n} \frac{1}{r} \sin\left(\frac{n\pi r}{r_s}\right) \exp\left(\frac{-Dn^2 \pi^2 r}{r_s^2}\right) \quad (A-1)$$

The rate of transfer at time t across the surface of the particle is:

$$4\pi r_s^2 N_{Ar}(t) = 4\pi r_s^2 D \left( \frac{\partial q_A}{\partial r} \right)_{r=r_s} \quad (\text{A-2})$$

The differential term on the right side can be obtained from Equation A-1, and thus

$$N_{Ar}(t) = + \frac{2r_s}{\pi r_s} D (q_{A0} - q_{A,t}^*) \sum_{n=1}^{\infty} \exp\left(\frac{-Dn^2 \pi^2 r}{r_s^2}\right) \quad (\text{A-3})$$

The total transfer up to time  $t$ ,  $N_A'$ , is as follows:

$$N_A' = 4\pi r_s^2 \int_0^t N_{Ar}(t) dt \quad (\text{A-4})$$

$$= 4\pi r_s^2 \int_0^t \frac{2}{r_s} D (q_{A0} - q_{A,t}^*) \sum_{n=1}^{\infty} \exp\left(\frac{-Dn^2 \pi^2 r}{r_s^2}\right) dt \quad (\text{A-5})$$

$$= 8\pi r_s D \int_0^t (q_{A0} - q_{A,t}^*) \sum_{n=1}^{\infty} \exp\left(\frac{-Dn^2 \pi^2 r}{r_s^2}\right) dt \quad (\text{A-6})$$

Likewise, the material balance of the transfer up to time  $t$ ,  $N_A'$ , is:

$$(q_{A0} - \bar{q}_A) \times (4/3)\pi r_s^3 = N_A' \quad (\text{A-7})$$

Rearranging,

$$\bar{q}_A = q_{A0} - \frac{3}{4\pi r_s^3} N_A' \quad (\text{A-8})$$

$$= q_{A0} - (q_{A0} - q_A^*) \left[ 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left(\frac{-Dn^2 \pi^2 t}{r_s^2}\right) \right] \quad (\text{A-9})$$

## Appendix 2

The surface diffusion equation for the boundary condition of Equation 7 is as follows.

$$D \left( \frac{\partial^2 C_r}{\partial r^2} + \frac{2}{r} \frac{\partial C_r}{\partial r} \right) = \frac{\partial C_r}{\partial t} \quad (\text{A-10})$$

Thus, the initial condition for Equation A-10 was the following relation.

$$D \left( \frac{\partial C_r}{\partial r} \right)_s = k_f (C - C_{r=rs}) \quad (\text{A-11})$$

By differentiating Equation A-11, we obtained Equation A-12.

$$D \left( \frac{\partial^2 C_r}{\partial r^2} \right)_s = k_f \left( \frac{\partial C_r}{\partial r} \right)_s \quad (\text{A-12})$$

Next, by substituting Equation A-12 into Equation A-10,

we obtained Equation A-13,

$$k_f \frac{\partial C_r}{\partial t} + D \frac{2}{r} \frac{\partial C_r}{\partial t} = \frac{\partial C_r}{\partial t} \quad (\text{A-13})$$

where the first term can be neglected because  $k_f \ll 2D/r$ .

$$\frac{\partial C_r}{\partial t} = D \frac{2}{r} \frac{\partial C_r}{\partial t} \quad (\text{A-14})$$

Thus, Equation A-10 becomes

$$\frac{\partial C_r}{\partial t} = \frac{2}{r} k_f (C - C_{r=rs}) \quad (\text{A-15})$$

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