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Dynamic Adsorption of Noble Gases

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Abstract—The breakthrough curves for radioactive gases passing through an adsorption bed are calculated using a modification of the Hougen-Marshall equation commonly used in chemical engineering. This model gives $\exp(-N)$ as a lower limit to the fractional reduction in activity of the gas passing through the bed, where N is the number of transfer units.

Chemical engineers, starting with Hougen and Marshall, 1947 (Ref. 1), often use the “J” function, described in detail in the *Chemical Engineer’s Handbook*,² for the modeling of adsorption beds to be used in the removal/concentration of components from a flowing stream.^{1,2} Burnette, Graham, and Morse, interested in the removal of radioactive krypton and xenon from a helium carrier gas, extended the “J” function to include radioactive decay.³ As follows, we give the equation of Burnette, Graham, and Morse and demonstrate its application to the dynamic adsorption of radon; we present this equation in the form in which it was originally given because it follows the terminology commonly used by chemical engineers:

$$\frac{c}{c_0} = e^{-\alpha x r / (1+r)} \left[1 - \frac{1}{1+r} e^{-\beta t (1+r)} \times \int_0^{\alpha x} e^{-\tau / (1+r)} I_0(2\sqrt{\beta \tau t}) d\tau \right], \quad (1)$$

where

C = concentration of adsorbate in the gas stream (mol/m³)

C_0 = gas phase concentration at inlet to adsorbent bed (mol/m³)

H = height of mass-transfer unit (m)

I_0 = modified Bessel function of the first kind and zero order (dimensionless)

K = bulk adsorption coefficient (m³/kg)

L = total length of adsorbent bed (m)

M = total mass of adsorbent in bed (kg)

$r = \lambda/\beta$

t = time (s)

Q = volume flow rate of gas (m³/s)

x = distance along adsorbent bed (m)

$\alpha = 1/H$ (m⁻¹)

$\beta = LQ/KHM$ (s⁻¹)

λ = radioactive decay constant (s⁻¹)

τ = dummy variable for integration (dimensionless).

The original equation of Hougen and Marshall can be recovered from Eq. (1) by setting $\lambda = 0$.

We have been looking at adsorbent beds to remove radon, a subject of interest to prior investigators.^{4–7} Here,

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we decided to apply the Burnette, Graham, and Morse equation to our analyses with the thought that the use of an equation consistent with current engineering practice would produce easily understandable results. Unexpectedly, we found that Burnette, Graham, and Morse's original equation can be rewritten in a simpler form:

$$\frac{C}{C_0} = \int_0^t \frac{N}{\sqrt{tt_0}} \exp(-\lambda t - N - Nt/t_0) I_1(2N\sqrt{t/t_0}) dt + \exp(-N), \quad (2)$$

where

N = number of transfer units = L/H (dimensionless)

t_0 = mean holdup time = KM/Q (s)

I_1 = modified Bessel function of the first order.

This form has the additional advantage of allowing us to calculate the breakthrough curve (i.e., the output from the bed as a function of time) with a single integration, whereas the original equation by Burnette, Graham, and Morse requires computing a new integral for each point on the breakthrough curve.

Technically, this new result comes from solving the partial differential equation for mass transfer using a Laplace transform of $t \Rightarrow s$ rather than the $x \Rightarrow s$ used earlier.

The time-based Laplace transform gives a simpler result because it is better suited to treat the time-dependent decay of radon.

The curves in Fig. 1 show the differences in the breakthrough curves for ^{220}Rn and ^{222}Rn . These curves show an interesting point, which is the second reason for this technical note. One might think that the shorter the isotopic half-life is, the lower the concentration of noble gas in the effluent is, but this is true only to a limited extent. It turns out that this equation predicts a nonzero output equal to $\exp(-N)$ at $t = 0$, no matter how short the isotopic half-life is. The reason for this somewhat surprising result comes from two basic assumptions in the original equation of Hougen and Marshall. The first assumption is in the definition of a transfer unit, namely, the average distance that an atom (or molecule) of an adsorbate must pass in the bed before it passes from the moving phase to the stationary phase. From this first assumption the fraction of adsorbate passing through the bed unadsorbed is equal to $\exp(-N)$. The second assumption is that there is no holdup in the moving phase. Thus, this unadsorbed fraction passes through the bed without any decay. This effect is shown clearly in Fig. 1, in the breakthrough curve for $n = 3$ transfer units, where the initial breakthrough for both ^{222}Rn and ^{220}Rn at $t = 0$ is 5% of the input, and is equally apparent in Eq. (2), which gives the term $\exp(-N)$ explicitly. These calculations

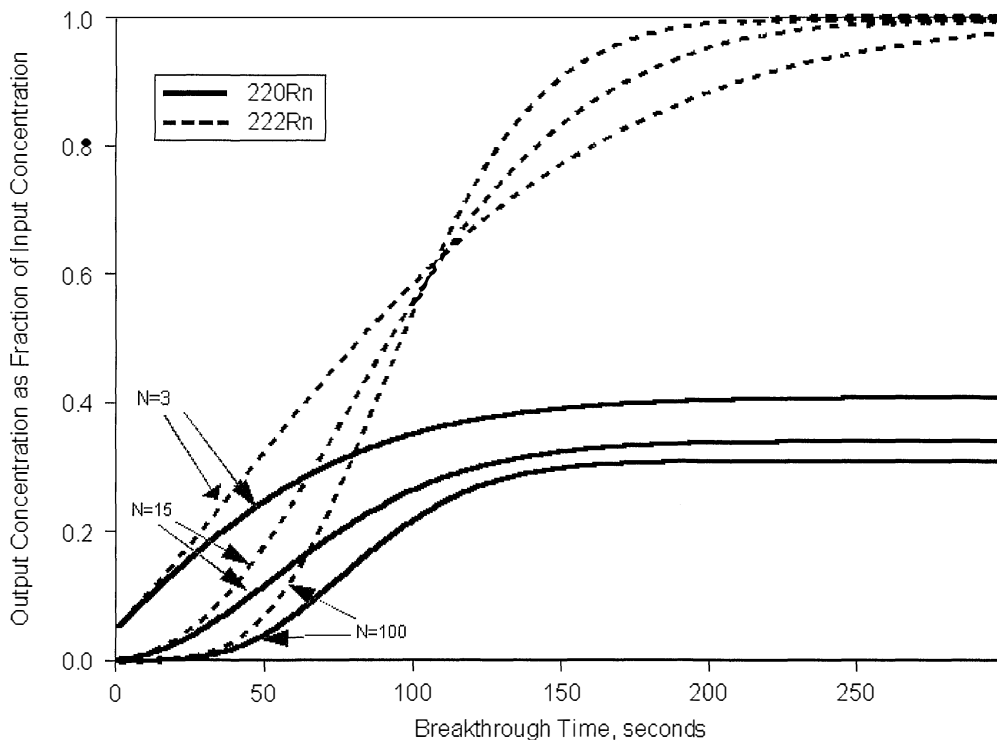


Fig. 1. Breakthrough curves for ^{220}Rn and ^{222}Rn . (Mean holdup time = 100 s; N = number of transfer units.)

can be extended to the removal of methyl iodide by impregnated activated carbon if the decay constant λ is set equal to the first-order rate constant for the reaction of methyl iodide with the impregnant.

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