LETTERS TO THE EDITOR

To the editor:

In a recent paper, Okazaki, Tamon and Toei (1981) presented a new hopping model for the surface transport of adsorbed gases. They supported the model with surfaces fluxes which were calculated from data on flow of adsorbing vapors through various porous materials. Some of these data had been obtained by the authors, and some came from the literature. In materials, the authors used the inverse-square-root-of-molecular-weight relationship, which may be stated as

$$|N_A^s| = |N_A^T| - |(M_B/M_A)^{1/2}N_B^g|. \quad (1)$$

(The absolute values are used because of the different types of experiment possible.)

As pointed out almost twenty years ago by Field et al. (1963), the use of Eq. (1) to calculate the surface flux assumes that "the adsorbed layer of gas on a pore wall does not affect the flow rate of gas through pore." After citing evidence that frequently this assumption may be invalid, Field and his coworkers warn that "calculated surface diffusion coefficients should be viewed with great caution until these effects can be more fully investigated." The soundness of this admonition was demonstrated by Bell (1971; Bell and Brown, 1973), who observed disparities between the results that he obtained experimentally and those predicted by the inverse-square-root relationship. Gas-adsorbate collisions were the ascribed cause for the difference in results, and further experimental studies by Spencer (1976; Spencer and Brown, 1975) and Wan (1977) confirmed the validity of Bell's diagnosis. Appropriate corrections for pore blockage were made in all cases. The experimental studies by these authors used porous adsorbents with both homogeneous and heterogeneous surfaces, so the effects are widespread. Modeling studies (Bell and Brown, 1974; Takur et al., 1980) showed that errors in calculated surface fluxes resulting from use of the inverse-square-root relationship are frequently serious, and sometimes drastic.

Insufficient details of their experimental results are presented by Okazaki and his coworkers for us to use the published techniques of modeling gas-adsorbent interactions to estimate the errors in their calculated surface fluxes. Some of the literature results they cited, however, have had such errors estimated. Three such studies are among those we evaluated (Thakur et al., 1980): 1) Ross's and Good's (1956) data on n-C_4H_{10} on Graphon; 2) the results of Horiguchi et al. (1971) for C_2H_6 on Vycor; and 3) those of Gilliland et al. (1974) for SO_2 on Vycor. In all three situations, including gas-adsorbate interactions in the calculation of the surface fluxes altered these calculated flux values significantly. The first two showed large differences, changing by a factor of about two. The error in the third was much less, displaying a difference of about 10%.

These results confirm that calculations of surface components of total fluxes must include consideration of gas-adsorbate interactions. Although clever experimental methods may render the effects of these interactions negligible (e.g., Lee and O'Connell, 1975), the insignificant quality of these interactions must first be justified before using the inverse-square-root relationship. This was not done by Okazaki and his coworkers. Thus, although their paper has made a valuable and praiseworthy addition to the conceptual modeling of surface-flow phenomena, the experimental support for their model must be regarded as uncertain.

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Notation

M_A = molecular weight of substance A, g/mol

M_B = molecular weight of substance B, g/mol

N_A^s = surface flux of substance A, mol/(cm^2)(s)

N_A^T = total flux of substance A, mol/(cm^2)(s)

N_B^g = flux of nonadsorbing substance B, mol/(cm^2)(s)

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Reply:

Our investigation (Okazaki et al., 1981) does not treat surface flow in the case where an adsorbed molecule would be expected to behave as a molecule in a two-dimensional gas. If the literature experimental data cited in our work show a two-dimensional gas behavior, we should exclude those data in verifying our hopping model for surface flow.

In our work, the counterdiffusions of nitrogen-helium, nitrogen-argon and argon-helium were measured. The diffusion behaviors showed the Knudsen flow and obeyed the inverse-square-root relationship within our experimental errors. Thus these components were regarded as nonadsorbable. As the diffusion rates of nitrogen in nitrogen-ethylene, nitrogen-propylene, nitrogen-isobutane and nitrogen-sulfur dioxide systems were observed and also estimated from the Knudsen equation by taking into account the corrections for pore blockage, the interactions between gas-adsorbate proposed by Brown et al. (1973, 1974, 1975 and 1980) would not be needed to be considered. However, we cannot make the above examinations for the literature data cited in our investigation because these data were obtained from permeability experiments.

We also reported surface self-diffusion data of sulfur dioxide on Vycor glass (Okazaki et al., 1981). The measured surface self-diffusion coefficient equals to the surface flow coefficient in the wide range of amount adsorbed. It seems to be difficult to understand this agreement from the standpoint of two-dimensional (mobile) gas behavior.

Bell and Brown (1974) and Thakur et al. (1980) proposed the models for gas-adsorbate collisional effects and verified these models using the observed diffusion behavior of helium from helium-nitrogen system and helium-propane system through pelleted Graphon. They regarded helium as nonadsorbable in their works. On the other hand, Horiguchi et al. (1971) experimentally suggested that the surface diffusion (surface flow) on Graphone took place even for helium gas at 0° to 110°C. We think that the investigations performed by Brown et al. are valuable and interesting, but it would need further experimental verifications.

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ERRATA

In "Drop Size and Continuous-Phase Mass Transfer in Agitated Vessels" by A. H. P. Skelland and Jai Moon Lee, [*AIChE J.*, **27**, 99 (1981)]:

Page 109, col. 2, 2nd paragraph, should read:

To obtain equal capacity coefficients in two geometrically similar units of different size, when $\phi_1 = \phi_2$ and the relevant physical properties have respectively the same values on the two scales, equating the right-hand side of Eq. 51 for systems 1 and 2 leads to

$$\frac{N_1}{N_2} = \left(\frac{d_{I_2}}{d_{I_1}}\right)^{0.966} \tag{60}$$

Equation (61) should read

$$\frac{P_1}{P_2} = \left(\frac{d_{I_1}}{d_{I_2}}\right)^5 \left(\frac{N_1}{N_2}\right)^3 = \left(\frac{d_{I_1}}{d_{I_2}}\right)^{2.102} \tag{61}$$

Equation (62) in corrected form is

$$\frac{P_1/Vol_1}{P_2/Vol_2} = \frac{P_1}{P_2}\left(\frac{d_{I_2}}{d_{I_1}}\right)^3 = \left(\frac{d_{I_2}}{d_{I_1}}\right)^{0.898} \tag{62}$$

The final paragraph should then read:

Eq. 62 shows that the power input per unit volume to make $(k_c a)_1 = (k_c a)_2$ decreases with increasing d_I for the range of d_I/T studied.

The paper "Modelling Highly Skewed Chromatographic Response Curves" by Gelbin et al., [*AIChE J.*, **28**, 177 (1982)] was published before the corrected galleys were received. The following is a table of corrections:

Eq.	Wrong	Right
(1)	$\left.\frac{\partial c}{\partial r_1}\right\|$	$\left.\frac{\partial c}{\partial r_1}\right\|_{r_1 = R_1}$
(3)		$c\|_{x=0} = c_\Delta \cdot t_0 \cdot \delta(t)$
(4)		$\lim_{x \to \infty} c(t,x) = 0$
(5)	Ba	B_a
(13)	Kai	K_{at}
(14)	$D_{modt}t$	$D_{mod,t}$
(15)	S	s
(16)	μ_{01emp}	$\mu_{0,emp}$
(17)	μ_1^1	μ'_1
(19)	K_{a1emp}	$K_{a,emp}$
(21)	κe_{emp}	κ_{emp}
(21)	μ_1^1	μ'_1
(23)	K_{a1emp}	$K_{a,emp}$
(24)	μ_{21emp}	$\mu_{2,emp}$
(26)	$\times$ (*mult.*)	X
(28)	D_{c1fr}	$D_{c,fr}$
(30)	ρ_t	ρ_t
Notation	$/u$	μ