# CORRESPONDENCE

## EFFECT OF GAS RATE ON OVER-ALL MASS TRANSFER COEFFICIENTS AND THE PERFECT MIXING THEORY

SIR: In a recent article, Buchanan, Teplitzky, and Oedjoe (*1*) discussed the performance of two types of gas-liquid contacting equipment—a vertically vibrated column and a sparger column—tested with a copper-catalyzed sulfite-air oxidation system. They arrived at no definite conclusions concerning the relative magnitudes of gas- and liquid-phase resistances, but did state that their results indicated substantial gas-film resistance. Some discussion of this claim is in order.

### Effect of Gas Rate on $K_Ga$

An increase in gas rate could increase measured $K_Ga$ values by:

Increasing gas-phase turbulence
Increasing interfacial area
Increasing liquid-phase turbulence which is due to the stirring action of the gas bubbles

It should be noted immediately that an increase in interfacial area would increase measured $K_Ga$ values regardless of which resistance controlled over-all mass transfer.

Consider now the three main types of gas-liquid contactors that have been investigated using the sulfite-air oxidation system and how an increase in gas rate can affect the operation of each.

### Gassed Agitated Tank

An increase in gas rate cannot significantly affect liquid turbulence, as this is predominantly due to stirring. At first glance, it might appear that increasing gas rate would increase interfacial area and gas phase turbulence, but in any case of interest this is not true.

### Sparger Column

An increase in gas rate does increase interfacial area, liquid turbulence due to the stirring action of gas bubbles, and, depending on the mechanism of bubble formation, can increase the internal turbulence of the newly formed bubbles.

### Vibrating Column

This apparatus corresponds in many respects to a shaken flask, bubbles being formed by submergence of gas in the violently agitated liquid and subsequently migrating in all directions through the liquid. An increase in gas rate could only affect turbulence in the vapor bulk above the liquid. This in turn might have some slight, but probably unimportant, effect on internal bubble turbulence. The interfacial area and liquid turbulence are functions only of equipment size, vibrational frequency, and amplitude. The gas-phase turbulence is in large part due to the action of the rapidly moving liquid. Since gas flow rates are small, no effect of gas rate on $K_Ga$ would be expected.

### Discussion

Buchanan, Teplitzky, and Oedjoe (*1*) state that their results for the variation of $K_Ga$ with $G$ for the sparger column agree with those of Cooper, Fernstrom, and Miller (*2*) for the gassed agitated tank. The experimental slope of the plot of log $K_Ga$ *vs.* log $G$ is 0.67 in both these cases. In 1957, however, Friedman and Lightfoot (*3*) pointed out that the data of Cooper, Fernstrom, and Miller could not be reproduced with respect to this slope and that their work indicated a zero dependence of $K_Ga$ on gas rate. A recent work of Westerterp, van Dierendonck, and de Kraa (*4*) has further demonstrated that above the minimum agitation speed there is no influence of $G$ on $K_Ga$ in a gassed agitated tank. The article further elucidated this independence by introducing the concept of "perfect mixing" in the gas phase.

Consider a well agitated tank being supplied with gas at a low rate via a sparger. Near and below the impeller, gas bubbles circulate rapidly in all directions, being continuously broken up and recoalescing. The rate of bubble circulation is high compared with the gas feed rate, and bubbles may remain in contact with the liquid for long periods before escaping the circulation pattern and rising to the surface. Further, the chance of a bubble escaping is only slightly, if at all, dependent on its age; a newly formed bubble has almost as high a probability of leaving after a few seconds as a bubble that has circulated for a much longer time. If one considers any arbitrary group of bubbles leaving the liquid, this will consist of bubbles having a spectrum of ages. In a well agitated system, this spectrum will be quite broad, and the exit gas composition will be that which is due to mixing of all these bubbles. If there is no relation between bubble age and probability of leaving the liquid, the resultant exit gas composition will represent an average of the gas contacting the liquid. In this case, the correct gas composition for evaluation of $K_Ga$ is not the log mean of the inlet and exit gas compositions, but the resultant exit gas composition. The criterion given for this type of behavior is that the rate of gas circulation due to the pumping action of the impeller be considerably higher than the rate of gas supply (*4*). Stated another way, the total gas holdup in the liquid at any time should be much greater than the total gas fed to the system in the time required for a bubble to rise vertically through the unstirred liquid.

Below the minimum agitation speed, gas rate does affect $K_Ga$, probably owing to an increase in interfacial area with gas rate and increased turbulence in either of the phases. Above the minimum agitation rate, interfacial area is essentially independent of gas rate, again because gas circulation rates are much higher than gas feed rates.

The fact that any average residence times for gas bubbles would be greater than the time required for decay of internal bubble turbulence (4 to 6 seconds) means that any effect of increased initial bubble turbulence will be negligible. The

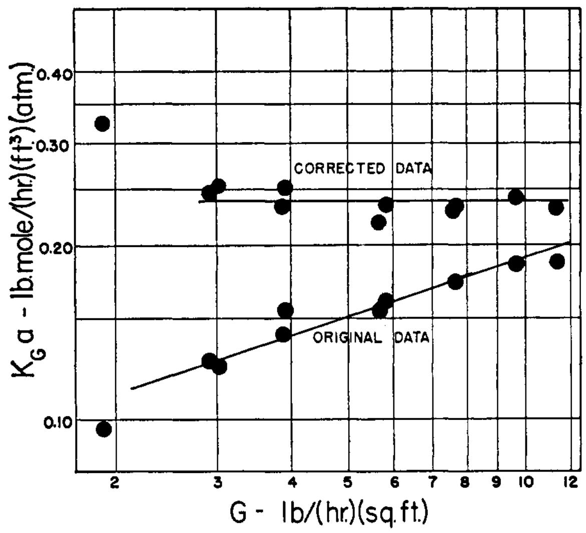


**Figure 1. Data of Buchanan recalculated using exit gas composition driving forces**

above discussion indicates that gas rate should not affect $K_Ga$ values in this type of contactor, as has been found experimentally (*3*, *4*). Westerterp and coworkers (*4*) also point out that the incorrect use of the log mean driving force leads to an apparent dependence of $K_Ga$ on $G$.

The sparger column is another case altogether. In this, gas simply bubbles through an unstirred column of liquid and an obvious relationship exists between bubble age and the probability of a bubble leaving the liquid. Thus, the exit gas cannot represent an average of the gas in contact with the liquid, and the use of a log mean driving force is indicated. The dependence on gas rate found by Buchanan and others may be due to an increase in interfacial area or a decrease in either film resistance through increased turbulence. Circulation decay within the bubbles would diminish any effect of increased initial gas turbulence in deep liquid columns, and for the case of a fast chemical reaction occurring within the liquid film, any liquid turbulence effects would be masked.

Under the conditions found in the vibrating column, as discussed previously, it would seem that there is no relationship between bubble age and the probability of a bubble leaving the liquid. The gas above the liquid is a well mixed (owing to liquid movement) composite of inlet gas and spent gas that has been in contact with liquid for various times. The same arguments apply here as in the agitated tank: the exit gas is a mixture of gas bubbles of all ages, and thus represents a good average of the gas contacting the liquid. In any event, the use of a log mean value of inlet and outlet gas compositions appears incorrect, since essentially none of the gas bubbles formed would have compositions the same as that of the inlet gas.

The data of Buchanan were recalculated on the basis of exit gas composition driving force to obtain new $K_Ga$ values. The calculations were necessarily quite approximate, as complete data were not available and the values used were obtained from Buchanan's Figures 3 and 4 (*1*). Figure 1 shows the results of these calculations. All values of $K_Ga$ lie at about the same level except that at the lowest gas rate. This point represents an oxygen efficiency of 87.5%, and for a gas phase concentration change of this magnitude, the use of such simple methods of calculation is of doubtful validity.

This zero dependence of $K_Ga$ on $G$ is to be expected if:

The interfacial area is independent of gas rate, as Buchanan stated it appeared to be

The gas phase flow has no effect on liquid phase turbulence, as is undoubtedly true

Either the gas phase resistance is not controlling or the average internal turbulence of the bubbles is not affected by gas rate

This last condition is fulfilled in the agitated tank because bubbles have long average "life spans" and circulation decay becomes important. In the vibrating column, gas rate would have little effect on internal bubble turbulence as bubbles are formed by submergence of parts of the bulk gas phase, which is only slightly agitated by gas feed rate.

### Conclusions

The vibrating column gas-liquid contactor should show no effect of gas rate on measured $K_Ga$ values. For well agitated systems, the use of the exit gas composition rather than the log mean driving force for evaluating mass transfer coefficients is indicated.

### Nomenclature

$a$ = interfacial area, sq. ft./cu. ft. volume
$G$ = gas rate, lb./(hr.) (sq. ft. cross section)
$K_G$ = over-all gas film mass transfer coefficient, lb. mole/(hr.) (sq. ft.) (atm.)

### Literature Cited


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*Robert G. Robinson*
*The Pennsylvania State University*
*University Park, Pa.*