

## A New Way of Enhancing Transport Process - The Hybrid Process Accompanied by Ultrafine Particles

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**Abstract**—A brief review is presented on the enhancing effects of fine particles on multiphase mass transfer in the first part of this paper. Some experimental results on the following aspects are shown and discussed: influence of fine particles on mass transfer parameters ( $k_L a$ ,  $k_L$ ,  $a$ ), enhancement due to particles adsorption and catalyzing a chemical reaction, and effects of particle size. The second part of this paper proposes two kinds of hybrid process accompanied by ultrafine particles: adsorptive distillation and slurry catalytic distillation. Finally, prospective fields for additional research are proposed.

Key words: Mass Transfer Enhancement, Enhancement Factor, Ultrafine Particles, Slurry System, Hybrid Process

### INTRODUCTION

Chemical engineering scientists and engineers have made their efforts for decades to enhance the transfer rate in several processes. The fine particles have been found to influence gas-liquid mass transfer parameters and especially enhance gas-liquid mass transfer for the particles that adsorb transferred component strongly or catalyze a chemical reaction involving a transferred component. The transport processes with slurry of fine particles are beneficial from the enhancement effects of fine particles on gas-liquid mass transfer. In general the hybrid process is a way of enhancing transport processes because it holds the advantages of both combined processes, and the shortcomings of one combined process are compensated by the other. In this paper, we introduce a new way of enhancing transport process, the hybrid process accompanied by ultrafine particles.

### ENHANCING EFFECTS OF FINE PARTICLES ON MULTIPHASE MASS TRANSFER

Gas-liquid slurry systems are often met in the chemical and petrochemical industries. For example, the fine particle catalyst of several microns in size is often used in slurry reactor, owing to its high effectiveness. Recently, more research work is focused on the enhancing effects of fine particles on multiphase mass transfer. We may consider the simplified mass transfer model in a gas-liquid slurry system, in which component A is transferred from gas phase to particles based on the film theory (Fig. 1).

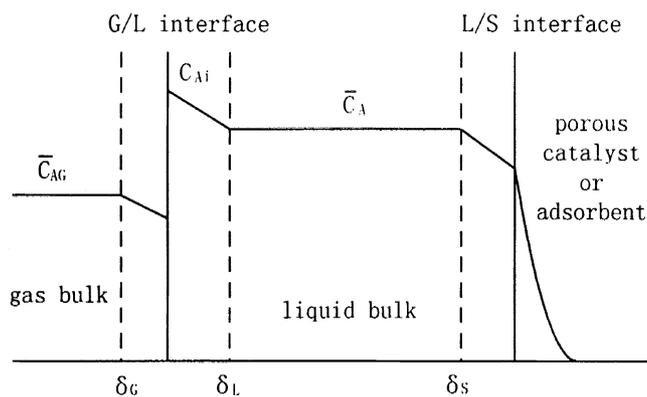
$$J_A a = \bar{c}_{AG} \left\{ \frac{1}{k_G a} + \frac{1}{m k_L a E_A} + \frac{1}{m k_S a_S} + \frac{1}{m k_P a_P} \right\} \quad (1)$$

$E_A$ , the enhancement factor of mass transfer at gas-liquid interface is defined as:

$$E_A = \frac{J_A \text{ with particles}}{J_A \text{ with the same but inert particles}} \quad (2)$$

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**Fig. 1. Concentration profiles for mass transfer into a slurry with catalyst or adsorbent particles.**

The presence of fine particles may influence the mass transfer, according to Eq. (1) and (2), in to ways:

- (1) influence on the mass transfer parameters;
- (2) enhancement of gas-liquid mass transfer by catalytic reactions and adsorption.

### 1. Influence of Presence of Fine Particles on the Mass Transport Parameters

1-1. Influence on the Volumetric Liquid-Side Mass Transfer Coefficient at the Gas-Liquid Interface ( $k_L a$ )

Since 1970's, much research has been carried out on the influence of fine particles on  $k_L a$ . Some interesting experimental results are shown in Fig. 2 and Fig. 3. Though there are no universal applicable relations predicting the influence of all types of particles in any liquid, it may conclude that in low solid hold-up,  $k_L a$  may increase remarkably for some types of particle and liquid. In high solid hold-up, it decreases causing by the increase of effective viscosity of slurry for most types of liquid.

1-2. Influence on the Gas-Liquid Specific Contact Area ( $a$ )

It is observed that there is an increase in interfacial area, with low

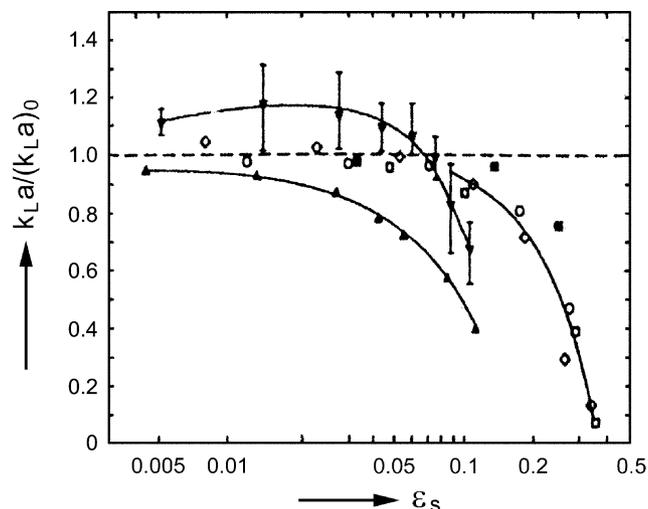


Fig. 2. Relative change in  $k_L \alpha$  in bubble column with respect to the particle-free system as a function of the solid volume fraction ( $\square$ ) PE-A (24.6  $\mu\text{m}$ )/ligroin; ( $\circ$ ) PE-B (106  $\mu\text{m}$ )/ligroin; ( $\diamond$ )  $\text{Al}_2\text{O}_3$  (10.5  $\mu\text{m}$ )/ligroin; ( $\nabla$ )  $\text{Al}_2\text{O}_3$  (10.5  $\mu\text{m}$ )/tetralin; ( $\blacksquare$ ) PE/Exsol after Michael and Reichert [1983] (from Öztürk and Schumpe, 1987).

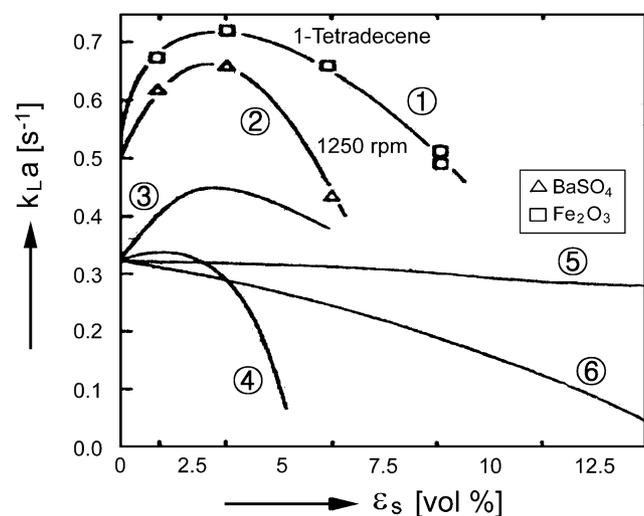


Fig. 3. Influence of solids on  $k_L \alpha$  in a 0.15 diameter stirred-tank reactor. (1)  $\text{Fe}_2\text{O}_3$  (<0.5  $\mu\text{m}$ ) in 1-tetradecene; (2)  $\text{BaSO}_4$  (<1  $\mu\text{m}$ ) in 1-tetradecene; (3)  $\text{Fe}_2\text{O}_3$  in 1,2,4-trimethyl benzene; (4)  $\text{BaSO}_4$  in 1,2,4-trimethyl benzene; (5) sea sand (<80  $\mu\text{m}$ ) in water; (6) kieselguhr (<50  $\mu\text{m}$ ) in water. Stirrer speed: 21 rps except for experiments with trimethyl benzene (16 rps) (composed from several Figures of Oguz et al., 1987).

solid hold-up for small particles, typically below 100  $\mu\text{m}$ . Some experimental results are listed in Table 1.

In low solid hold-up, the small particles may cover the bubble surface, thus preventing coalescence of the bubbles and resulting in smaller bubbles and an increased specific surface. The effect of an increase in  $\alpha$  is enhanced further by the fact that smaller bubbles have lower rising velocities, which leads to increasing gas hold-up,  $\epsilon_g$ . However, adding more fine particles (typically above 0.6

Table 1. Enhancement factor in different slurry systems

| Particle                                 | Solid content | Increase factor of $\alpha$ | Investigator                |
|--|---------------|-----------------------------|-----------------------------|
| Fines                                    | 0.3 wt%       | 200%                        | Sharma and Mashelkar [1968] |
| $\text{CaCO}_3$                          | 0.6 wt%       | 70%                         | Joshi and Sharma [1976]     |
| 7 $\mu\text{m}$ $\text{Ca}(\text{OH})_2$ | $\leq 25$ wt% | $\leq 25\%$                 | Sada et al. [1983]          |

vol%) can result in a decrease, possibly because the extra particles cannot find any uncovered bubble surface left and have to remain in the bulk, so contributing to an increased effective viscosity of slurry.

1-3. Influence on the Liquid-Side Mass Transfer Coefficient at the Gas-Liquid Interface ( $k_L$ )

Fine particles may affect  $k_L$  in tow ways. Firstly, the presence of fine particles close to the interface may reduce the effective fraction of liquid available for diffusion, by which,  $k_L$  can be reduced via the lowering of the effective diffusivity  $D_A$ . Secondly, the particles may influence the hydrodynamics close to the gas-liquid interface, thus affecting  $k_L$  via changing film thickness  $\delta_L$  and contact time  $t_c$ .

In the case of small fraction of heavy particles, as showed in Fig. 4, a moderate enhancement up to 30% seems possible depending on factors not yet well understood.

## 2. Enhancement of Gas-Liquid Mass Transfer Due to Presence of Fine Particles

Fing particle may enhance the gas-liquid mass transfer by several ways. In this paper, only tow types of enhancement are dealt with, they are:

- (1) Particles in the mass transfer zone adsorb the transferred gas

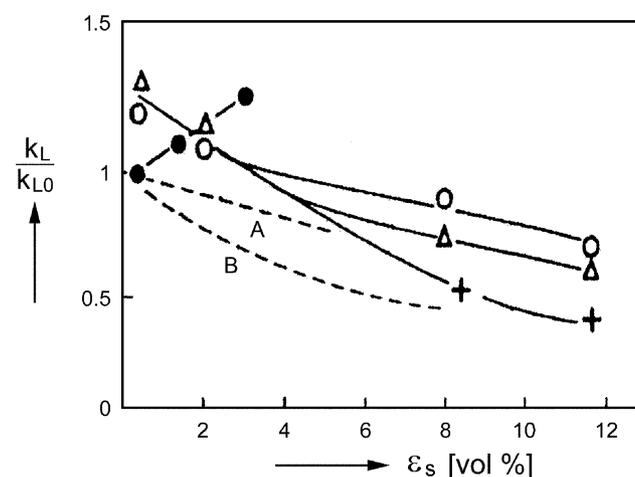


Fig. 4. The influence of small heavy particles on the mass transfer coefficient in small bubble columns: (—) aqueous solutions of 0.8 M  $\text{Na}_2\text{SO}_4$ . ( $\bullet$ ) Alumina,  $d_p=8 \mu\text{m}$ ,  $\rho_p=3,180 \text{ kg/m}^3$ ,  $2 \leq u_g \leq 6 \text{ cm/s}$ ; open symbols: kieselguhr,  $d_p=6.6 \mu\text{m}$ ,  $\rho_p=2,360 \text{ kg/m}^3$ ; ( $\circ$ )  $u_g=6 \text{ cm/s}$ , ( $\triangle$ )  $u_g=4 \text{ cm/s}$ , (+)  $u_g=2 \text{ cm/s}$ . Dashed curves: aqueous solution of 0.2 M NaOH: (A) calcium hydroxide,  $d_p=7 \mu\text{m}$ ,  $\rho_p=2,240 \text{ kg/m}^3$ ; (B) magnesium hydroxide,  $d_p=2 \mu\text{m}$ ,  $\rho_p=2,360 \text{ kg/m}^3$  (from Schumpe et al., 1987 and Sada et al., 1983).

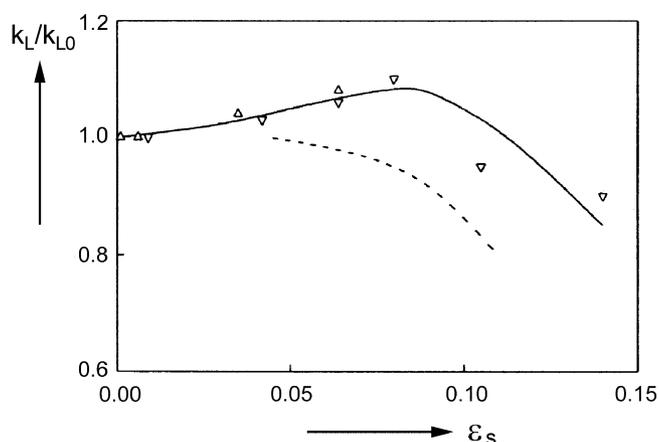


Fig. 5. Influence of inert solids of  $k_L$  in a stirred cell: (—) aqueous 0.5 M  $\text{NaHCO}_3$ /0.5 M  $\text{Na}_2\text{CO}_3$ , ( $\nabla$ ) 5-10  $\mu\text{m}$  kieselguhr, Quicker et al. [1989], ( $\Delta$ ) 5-10  $\mu\text{m}$  alumina, Quicker et al. [1989]. (—) Fine (10  $\mu\text{m}$ )  $\text{Ca}(\text{OH})_2$  particles. Sada et al. [1984].

phase component only physically (also known as the shuttle mechanism or grazing particles);

(2) Particles in the mass transfer zone catalyze a chemical reaction involving the transferred gas phase component.

#### 2-1. Enhancement Due to Adsorption of the Transferred Component on Particles

In a gas-slurry system, the gas phase containing pure A is absorbed into slurry of fine particles of about several microns, its size should be less than the liquid film thickness, and the component A is adsorbed strongly on the fine particles. The enhancement of gas-liquid mass transfer of component A, is due to the adsorption of A on the particles in the mass transfer zone close to the gas-liquid interface and the subsequent desorption and redistribution of A in the

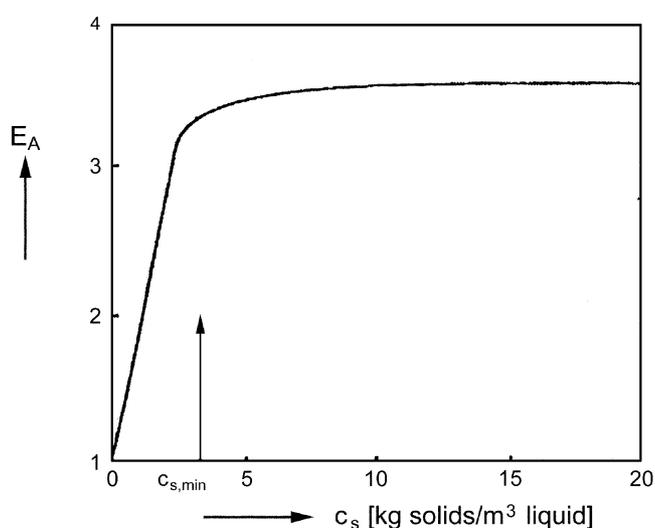


Fig. 6. Enhancement factor with physical absorption of  $\text{CO}_2$  in aqueous solutions of activated carbon particles adsorbing  $\text{CO}_2$ . Flat in interface stirred cell,  $\omega=1.33 \text{ s}^{-1}$ ,  $d_p < 5 \mu\text{m}$ .  $c_{s,min}$  is the solids concentration where the enhancement levels off (from Alper et al., 1980).

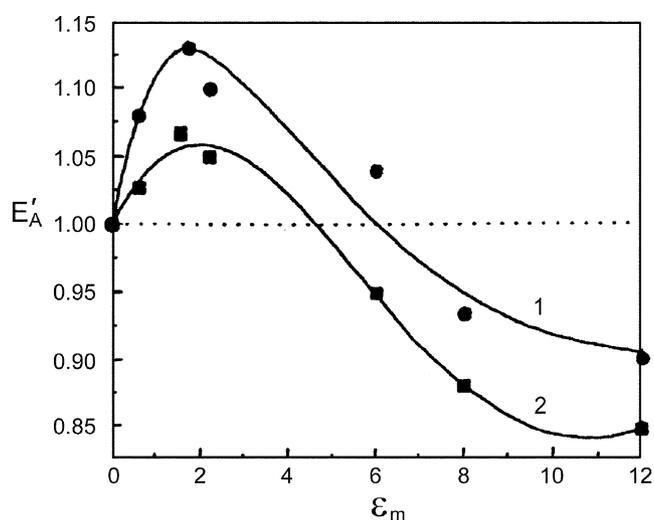


Fig. 7. Enhancement factor in bubble column with physical absorption of methanol in methanol-aceton-13X zeolite system.  $d_p \approx 10 \mu\text{m}$ . 1.  $u_G=3 \text{ cm}\cdot\text{s}^{-1}$ ; 2.  $u_G=5 \text{ cm}\cdot\text{s}^{-1}$  (from Cheng et al., 1999).

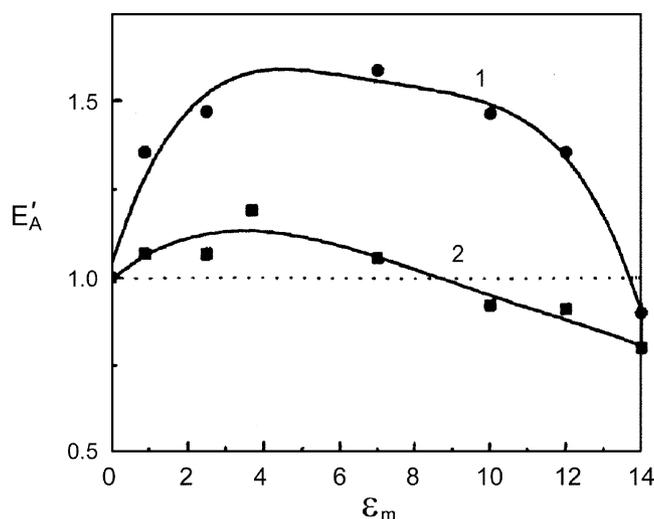


Fig. 8. Enhancement factor in bubble column with physical absorption of methanol in methanol-acetone-4A zeolite system.  $d_p \approx 10 \mu\text{m}$ . 1.  $u_G=5 \text{ cm}\cdot\text{s}^{-1}$ ; 2.  $u_G=3 \text{ cm}\cdot\text{s}^{-1}$  (from Cheng et al., 1999).

liquid bulk (the so-called grazing effect, first discovered by Kars and Best [1979]).

Enhancement of gas absorption in slurry due to its adsorption on fine particles has been observed by many authors. Some results are given in Fig. 6 to Fig. 8. In Fig. 6,  $E_A$  is defined by Eq. (2). While in Fig. 7 and Fig. 8, the enhancement factor of gas-liquid mass transfer of component A,  $E'_A$  is defined as:

$$E'_A = \frac{J_A \text{ with adsorbent particles}}{J_A \text{ without adsorbent particles}} \quad (3)$$

This enhancement factor  $E'_A$  consists of two contributions: the contribution of influence of gas-liquid mass transfer parameters by adding particles and the contribution of adsorption of component

A on particles, and the latter is  $E_A$  defined by Eq. (2).

Under some assumptions, the following expression for  $E_{A,max}$  is obtained [Vinke, 1992]:

$$E_{A,max} = \frac{4D_A}{k_L d_p} \tag{4}$$

For  $k_L$  in aqueous systems typically  $10^{-4}$  m/s,  $D_A$  typically  $10^{-9}$  m<sup>2</sup>/s and  $d_p=5 \mu\text{m}$ , we have  $E_{A,max}=8$ , which is an important value for industrial applications. We note also that if finer particles are used the enhancement factors will increase.

2-2. Enhancement Due to Particles Catalyzing a Chemical Reaction Involving the Transferred Component

If the catalyst particles have a diameter much smaller than the thickness of the mass transfer film and if sufficient particles are available in the film, then enhancement of gas absorption due to chemical reaction may occur, provided the specific chemical conversion rate is high enough. Chemically enhanced gas-liquid mass transfer with fine catalyst particles in slurry has been observed by several authors and for several chemical systems.

In Fig. 9, Rh/Al<sub>2</sub>O<sub>3</sub> particles gave no enhancement, but the Pd/C particles did even at relative low bulk particles loadings. Similar results can be obtained from Pd/Al<sub>2</sub>O<sub>3</sub> and Pd/AcC particles in Fig. 10; here  $E_A^0$  is defined as  $J_A/k_L C_{A^*}$ . The hydrophobic particles in hydrophilic liquids tend to stick to the gas-liquid interface, and thus may realize a significant enhancement. The enhancement of gas-liquid mass transfer in slurries by catalytic particles is an interesting phenomenon, which can be of practical importance if particles accumulate at the gas-liquid interface.

3. Effects of Particles Size on Multiphase Mass Transfer

From Fig. 2 to Fig. 4, we can see that in low particle hold-up region the fine particles have a positive influence on gas-liquid mass transfer parameters, and the finer the particles are, the stronger the

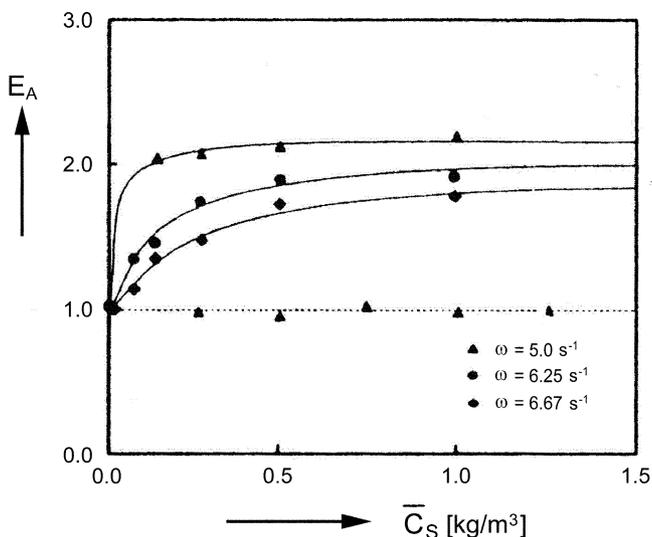


Fig. 9. Enhancement factor measured as a function of catalyst particle concentration in the suspension at different stirrer speeds. Dashed curve: Rh/Al<sub>2</sub>O<sub>3</sub>-demi-water-hydrogen system,  $\omega=6.67 \text{ s}^{-1}$ . No particle-to-bubble adhesion occurs. Other curves: Pd/C-demi-water-hydrogen system,  $\omega$  ranged from 5.0 to 6.67 s<sup>-1</sup>, Particle-to-bubble adhesion occurs (from Vinke, 1992).

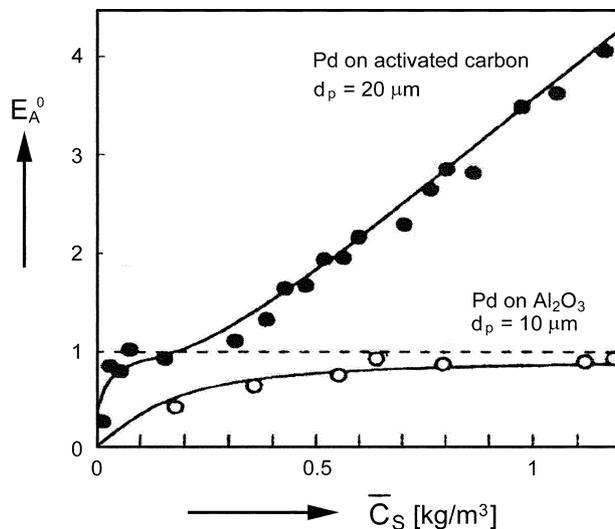


Fig. 10. Enhancement of absorption of hydrogen into aqueous hydroxylamine phosphate solutions by catalyst particles as a function of the catalyst loading of the bulk; stirred-tank reactor. Drawn lines are model calculations with best-fit parameters (from Wimmers and Fortuin 1988a).

influence is.

If the fine particles are assumed spherical, the specific liquid-solid interface as Eq. (5) can be obtained.

$$a_s = 6c_s / (\rho_p d_p) \tag{5}$$

For a fixed concentration of solid particles  $c_s$ ,  $\alpha_s$  increases directly with the decrease of particles size,  $d_p$  and thus benefits the liquid-solid mass transfer.

It has been shown by the research of enhancement mechanism of gas-liquid mass transfer due to particle adsorption or catalyzing reactions that enhancement does take place only if the particle's di-

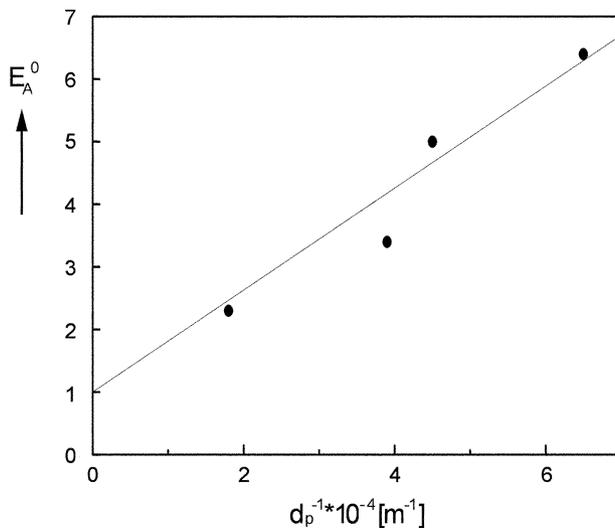


Fig. 11. Enhancement factor for hydrogen absorption in an aqueous hydroxylamine solution as a function of the reciprocal Sauter mean diameter of Pd/activated carbon catalyst particles,  $c_s=1.5 \text{ kg/m}^3$ , fiat interface stirred cell (from Wimmers et al., 1988).

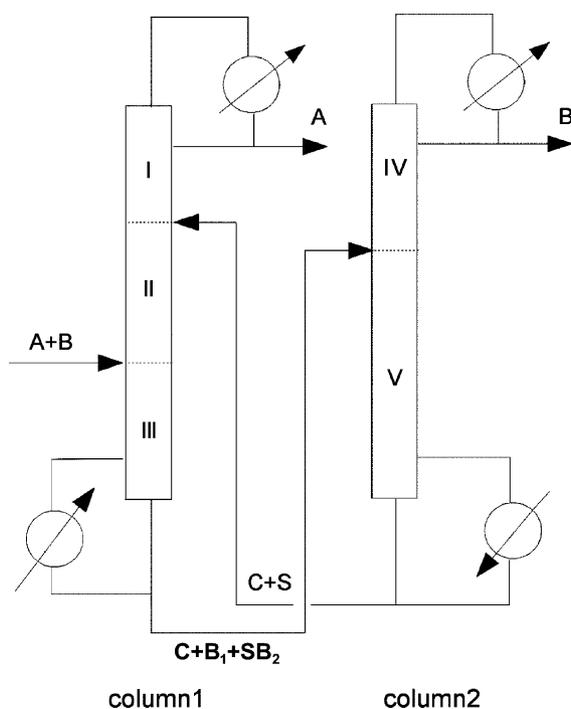
ameter is less than the thickness of the liquid side film at the gas-liquid interface. From Eq. (4),  $E_{A,max}$  increases with the decrease of particle diameter. As a result, the finer adsorbent or catalytic particles have more significant enhancement effect on gas-liquid mass transfer. An example of influence of catalytic particle size on the enhancement of gas-liquid mass transfer is shown in Fig. 11.

It is obvious that the finer particles have stronger enhancing effect on multiphase mass transfer; unfortunately, most of the research work on enhancing effects of fine particles on multiphase mass transfer is limited in the region of particle size larger than several microns. It leaves enormous challenges to explore: enhancing effects and mechanisms of ultrafine particles of size less than  $1\ \mu\text{m}$ ; researching and developing new processes by using slurries of ultrafine particles.

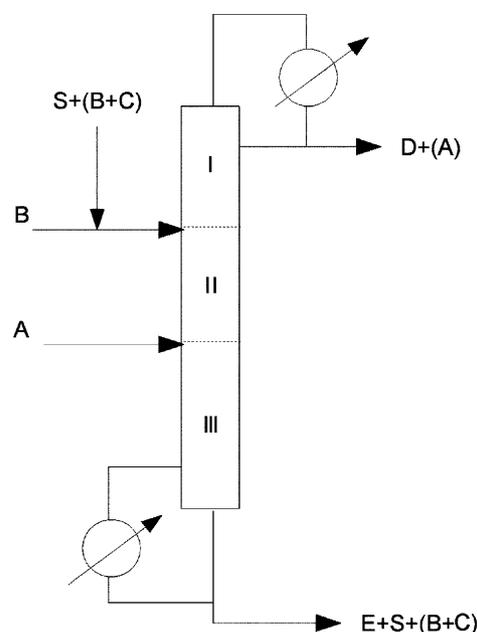
### HYBRID PROCESSES ACCOMPANIED BY ULTRAFINE PARTICLES

The hybrid process is usually more effective than the ordinary process. It is beneficial since the shortcomings of one process can be compensated by the other, and it has the advantages of both combined processes. For example, reactive distillation is a well known hybrid process, in which the separation factors of distillation is enhanced by reaction and/or the reaction conversion ratio is enhanced by distillation, and it has been found more and more application in industries.

We may expect higher enhancing effect if ultrafine particles are introduced to hybrid transport processes, such as reactive distilla-



**Fig. 12. Schematic flow sheet of adsorptive distillation. I, component A (more volatile) enriching section; II, adsorptive distillation section; III, component A stripping section; IV, component B enriching section; V, desorption (component B stripping) section.**



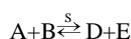
**Fig. 13. Schematic flowsheet of slurry catalytic distillation. I, component D (more volatile) enriching section; II, reactive distillation section; III, component A stripping section.**

tion. This new type of process undergoes double enhancing actions: enhancement of multiphase mass transfer and enhancement of combined process. Two hybrid processes accompanied by ultrafine particles are briefly described below: adsorptive distillation and slurry catalytic distillation. The flowsheets of the two processes are shown in Figs. 12 and 13, respectively.

The adsorptive distillation processes was first proposed in 1995 [Zhou et al., 1995], in which the fine particles of molecular sieve in size of several microns were used as adsorbent. The fine particles, S, used in adsorptive distillation are adsorbent with high adsorption selectivity for less volatile component B in the key separation component pair. An inert and the least volatile component C is used as liquid carrier of fine particles and forms the slurry, which is easily pumped between two columns. The hybrid operation of adsorption and distillation takes place simultaneously in column 1, increasing the separation factor, and then comes another hybrid operation of desorption and distillation in column 2, enhancing the regeneration of the adsorbent. The fine adsorbent particles may also enhance the mass transfer during the whole operation. Thus the adsorptive distillation process holds the advantages of high separation factors, high mass transfer rate, low energy-consumption and capability of large-scale production. It is especially suitable to the separation of azeotropic and isomeric systems as well as to those requiring high product purity. It was successfully used to obtain pure ethanol from ethanol-water mixture and removal of aromatics from light oil. The separations of pure iso-propanol and tertiary butanol from their aqueous solutions are now undertaken by using ultrafine adsorbent. We may expect the adsorptive distillation to be more effective and applicable if ultrafine adsorbent particles are used. As mentioned above, ultrafine adsorbent particles ( $\leq 1\ \mu\text{m}$ ) have stronger enhancing effects of gas-liquid mass transfer, lower liquid-solid and inter-particles mass transfer resistance (compared with fine particles in size of sev-

eral microns in gas-slurry system). The slurry formed by ultrafine particles poses no problem of precipitation of solid in process equipment.

Slurry catalytic distillation is a new kind of heterogeneous reactive distillation process in which the ultrafine particles are used as catalyst and with liquid form the slurry. The vapor phase and liquid slurry phase contact countercurrently in a distillation column. Unlike adsorptive distillation, the flowsheets of slurry catalytic distillation may differ from each other depending on the reaction type and the relative volatility of reactants and products. The inert carrier liquid C is not always necessary for some cases, the excess of one reactant, for even a part of product may be used as carrier liquid to form catalyst slurry recycled between the two columns. In general, the hybrid operation of reaction and distillation takes place in section II in reactive distillation column. We consider a general slurry catalytic distillation process with chemical reaction:



The more volatile product D is withdrawn from top of the column and the less volatile product E with catalytic slurry is pumped from the bottom to a distillation column in which the product E is removed and recovered. The recovered catalytic slurry from the distillation column is recycled to the reactive distillation column.

Traditional heterogeneous reactive distillation processes are carried out in two types of column: tray columns in which catalyst packages are stacked on tray, and packing columns in which catalyst is coated on the surface of the packing. Compared to traditional heterogeneous reactive distillation columns, the slurry catalytic distillation column is simpler in structure and lower in loading of catalyst and investment.

The ultrafine catalyst particles may also enhance the vapor-liquid, liquid-liquid, and inter-particles mass transfer during the whole process. Thus the slurry catalytic distillation process possesses the advantages of high reaction conversion ratio or high separation factor, high mass transfer rate, low energy-consumption and capability of large-scale production. Some experimental works of slurry catalytic distillation process are being carried out, such as alkylation of benzene, hydration of olefins and esterification of organic acid.

## CONCLUSION AND PROSPECTS

By using a slurry of ultrafine particles of adsorbent or catalyst, the adsorptive distillation and slurry catalytic distillation are new kinds of hybrid process. Their advantages are that they:

1. Possess advantages and overcome shortcomings of both combined processes.
2. Have higher enhancing effects of gas-liquid mass transfer because of the addition of ultrafine adsorbent or catalyst.
3. Have lower liquid-solid and intraparticle mass transfer resistances.
4. Eliminate the trouble of precipitation of solid in pipe and device.
5. Have simpler column construction and lower equipment investment.

These new hybrid processes should be more effective and appli-

cable. If ultrafine particles are used as adsorbent or catalyst in the hybrid processes mentioned above, the following aspects are to be of interest in research:

- Influence of ultrafine particles on slurry transport properties.
- Enhancement factor of ultrafine adsorbent or ultrafine catalyst on gas-liquid mass transfer in slurries.
- Hybrid processes accompanied by ultrafine particles and their applications.

Recently, material science and technology are rapidly developing and many nano-materials including even catalysts and adsorbents occur. The particle size of nano-materials is less than 100 nm. They possess very high specific surface area. In a slurry of such particles, the interaction between particles and liquid molecules becomes stronger and is no longer negligible. The adsorbent or catalyst of nano-materials possesses more active sites on the particle surface. Can nano-particles be used as adsorbent or catalyst in the hybrid processes mentioned above? What happens if these nano-particles are used in hybrid processes? We hope that our research work may answer these questions in the near future.

## ACKNOWLEDGMENTS

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## NOMENCLATURE

|             |   |
|-------------|---|
| $a$         | : specific gas-liquid contact area [ $\text{m}^2/\text{m}^3$ ]  |
| $a_p$       | : specific external interface of solids [ $\text{m}^2/\text{m}^3$ ]   |
| $a_s$       | : specific internal interface of solids per unit porous particle volume [ $\text{m}^2/\text{m}^3$ ]                 |
| $\bar{c}_A$ | : bulk concentration of species A [ $\text{kmol}/\text{m}^3$ ]  |
| $c_{Ai}$    | : concentration of species A in liquid at gas-liquid interface [ $\text{kmol}/\text{m}^3$ liquid]                   |
| $C_s$       | : concentration of solids in liquid [ $\text{kg solids}/\text{m}^3$ liquid]   |
| $C_{s,min}$ | : minimum concentration of solids for which maximum enhancement is obtained [ $\text{kg solids}/\text{m}^3$ liquid] |
| $d_b$       | : bubble diameter [m]   |
| $d_p$       | : particle diameter [m]   |
| $D$         | : molecular diffusion coefficient [ $\text{m}^2/\text{s}$ ]   |
| $E_A$       | : enhancement factor of component A, defined by Eq. (2), dimensionless  |
| $E_A^0$     | : $J_A/k_L c_{Ai}$ , dimensionless  |
| $E_{A,max}$ | : maximum enhancement factor, dimensionless   |
| $E'_A$      | : enhancement factor of component A, defined by Eq. (3), dimensionless  |
| $J_A$       | : molar flux of A [ $\text{kmol}/(\text{m}^2 \cdot \text{s})$ ]   |
| $k_G$       | : gas-liquid mass transfer coefficient [m/s]  |
| $k_L$       | : liquid-side mass transfer coefficient [m/s]   |
| $k_p$       | : overall particle adsorption rate constant [m/s]   |
| $k_s$       | : liquid-to-solid mass transfer coefficient [m/s]   |
| $m$         | : gas solubility, $c_L/c_G$ at equilibrium  |
| $t$         | : time [s]  |
| $t_c$       | : contact time in penetration model [s]   |
| $u$         | : superficial velocity [m/s]  |

- $\delta_l$  : film thickness in terms of the film model ( $=D/k_f$ ) [m]  
 $\delta_G$  : gas film thickness in terms of the film model [m]  
 $\delta_s$  : film thickness in terms of the film model for mass transfer to the particles [m]  
 $\varepsilon_G$  : gas hold-up [ $\text{m}^3/\text{m}^3$  liquid]  
 $\varepsilon_m$  : mass solid hold-up [kg solid/kg liquid]  
 $\varepsilon_s$  : solid hold-up [ $\text{m}^3/\text{m}^3$  liquid]  
 $\rho_p$  : particle density [ $\text{kg}/\text{m}^3$  solids]  
 $\omega$  : stirring speed [ $\text{s}^{-1}$ ]

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