PERGAMON

Chemical Engineering Science 56 (2001) 621–626

Chemical Engineering Science

www.elsevier.nl/locate/ces

Modelling and simulation of 1,2-dichloroethane production by ethylene oxychlorination in fluidized-bed reactor

S. M. Al-Zahrani[a,*], A. M. Aljodai[b], K. M. Wagialla[a]

[a]*Chemical Engineering Department, King Saud University, P.O. Box 800, Riyadh 11421, Saudi Arabia*
[b]*SABIC R&T, P.O. Box 42503, Riyadh 11551, Saudi Arabia*

Abstract

A comprehensive reactor model for ethylene oxychlorination for the production of 1,2-dichloroethane in a fluidized-bed reactor is developed. The model is based on the two-phase theory of fluidization and allows for the change in volumetric gas flow rate in the dense phase due to the change in number of moles accompanying the reaction. The model predictions compared favorably with the industrial data obtained from the literature. The effect of different parameters on the behavior of the system is also investigated. It has been found that the bubble diameter, ethylene molar feed fraction, residence time, and height at minimum fluidization have significant effects on the reactor performance. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Dichloroethane; Oxychlorination; Ethylene; Fluidized-bed; Modelling; Simulation

1. Introduction

Vinyl chloride monomer (VCM) or ($CH_2 = CHCl$) is the basic feed for the production of polyvinyl chloride (PVC), which is one of the most massively produced thermoplastics in the chemical industry. The current worldwide VCM production capacity is about 24 MM ton per year (SRI report, 1998). VCM is produced commercially by cracking of 1,2 dichloroethane ($CH_2CH_2Cl_2$ or EDC) which is manufactured worldwide by either the direct chlorination or oxychlorination of ethylene. Around 85% of the total EDC production is used for the production of vinyl chloride. 10% is used in the production of chlorinated solvents such as 1,1,1-trichloroethane and tri- and tetrachloroethylene. The rest goes into various processes, mainly for synthesis of ethylenediamines (Nawroski & Velez, 1983).

In commercial ethylene oxychlorination reactors, gaseous ethylene, HCl, and air (or oxygen) catalytically react at temperatures in excess of 200°C to produce EDC. The overall chemical reaction is

$$C_2H_4 + 0.5O_2 + 2HCl \xrightarrow{CuCl_2} CH_2CH_2Cl_2 + H_2O,$$

$$\Delta H_r^0 = -295\ \text{kJ/mol}$$

Typically, the catalyst is composed of cupric chloride supported on high-surface-area alumina. Although other supports such as graphite, silica gel, calcined Fuller's earth, diatomaceous earth, pumice, or kieselguhr may be used, alumina is generally preferred because of its performance, attrition resistance and the ability to control its surface area (Ullmann, 1986). Other metal salts, such as potassium, sodium, or aluminum chloride, may be added to the catalyst to increase selectivity and reduce volatilization of the copper chloride (Ullmann, 1986).

Because the oxychlorination reaction is highly exothermic, substantial quantities of heat must be removed from the reactors. Fluidized-bed reactors have many advantages in this system. Near-isothermal operation can easily be achieved with a well-fluidized catalyst, giving an overall heat-transfer coefficient to the cooling surface in the range of 250–500 W/m^2K (Smallwood, Stephenson, Newman & Bunten, 1987).

In fluidized beds, the heat is removed by internal cooling coils that are submerged in the fluid bed (Smallwood, Stephenson, Newman & Bunten, 1987). Reaction temperatures are generally controlled in the range 210–240°C.

* Corresponding author. Tel.: +00966-1-4676873; fax: +00966-1-4678-770.

E-mail address: szahrani@ksu.edu.sa (S. M. Al-Zahrani).

0009-2509/01/$ - see front matter
PII: S0009-2509(00)00268-2

Because the fluid bed is essentially isothermal, reaction conditions are uniform throughout the bed. An optimum reactor temperature can be achieved by proper design and operation of the heat removal system. Fluidized-bed catalysts must have good structural integrity so that excessive amounts of fines are not generated. Care must be taken to avoid sticky catalyst particles because catalyst agglomeration can lead to poor fluidization or loss of fluidization entirely (Ullmann, 1986).

In an air-based oxychlorination process, ethylene is usually fed in a slight stoichiometric excess relative to HCl (1–4% molar), but air may be in substantial excess (20–50% molar). The objective is to seek high HCl conversion ($> 98\%$) but to minimize the loss of ethylene to vent. The use of oxygen permits a more flexible policy. Here it is customary to recycle the major part of the gases after EDC and water removal, so it is possible to operate with a higher amount of ethylene fed to the reactor. This gives better HCl conversion and lower organic by-products in the EDC as well as lower losses of unconverted ethylene (Keane, Stobaugh & Townsend, 1973; Wimer & Feathers, 1976). Frequently, this purge may be vented to the atmosphere or, if incineration is required, the size of the incineration plank and the amount of support fuel used are much less than for air-based oxychlorinations (Wimer & Feathers, 1976; Reich, 1976; Markeloff, 1984).

The purpose of this study is to develop a mathematical model to describe the ethylene oxychlorination reaction in a fluidized-bed reactor. In the proposed study, the rigorous theory of the two-phase flow was used to model the fluidized-bed reactor. The effects of operating and design parameters were studied. The parameters include effect of feed mole fractions, residence time, bubble diameter, particle diameter, feed temperature, cooling medium temperature, pressure, height at minimum fluidization (H_{mf}), velocity at minimum fluidization (U_{mf}) on the reactor performance. The present model was validated using a real plant data reported in the literature.

Table 1
The hydrodynamic, transport and physical parameters (Aljodai, 1999)

Parameter	Theoretical or empirical expression
Bed voidage at minimum fluidization	$\varepsilon_{mf} = 0.586\phi^{-0.72}\left(\frac{\mu^2}{\rho_g g(\rho_s-\rho_g)d_p^3}\right)^{0.029}\left(\frac{\rho_g}{\rho_s}\right)^{0.021}$
Superficial velocity at minimum fluidization	$\frac{1.75}{\varepsilon_{mf}^3}Re_m^2 + \frac{150(1-\varepsilon_{mf})}{\varepsilon_{mf}^3}Re_m - \frac{d_p^3\rho_g(\rho_s-\rho_g)g}{\mu^2} = 0$ $Re_{mf} = \frac{\rho.D_p.U_{mf}}{\mu}$
Bubble diameter	$d_B = d_{BM} - (d_{BM} - d_{Bo})e^{-0.3h/D}$ $d_{BM} = 0.652[A(U_o - U_{mf})]^{0.4}$ $d_{Bo} = 0.00376(U_o - U_{mf})^2$
Bubble rising velocity	$U_b = U_o - U_{mf} + 0.711(gd_B)^{0.5}$
Fraction of bubble phase	$\delta = \frac{U_o - U_{mf}}{U_b}$
Coefficient for mass transfer	$\frac{1}{K_{be}} = \left(\frac{1}{K_b} + \frac{1}{K_e}\right)$ $K_b = 4.5\frac{U_{mf}}{d_B} + 10.4\frac{D_e^{0.5}}{d_B^{1.25}}$ $K_e = 6.78\left(\frac{\varepsilon_{mf}DU_b}{d_B^3}\right)^{0.5}$
Coefficient for heat transfer	$H_{bd} = 4.5\left(\frac{U_{mf}\rho_g C_{pg}}{d_B}\right) + 10.4\left(\frac{\rho_g k_g C_{pg}}{d_B^{2.5}}\right)^{0.5}$
Heat transfer coefficient	$h_w = 0.88\frac{k_g}{d_p}\left(\frac{\rho_g(\rho_s-\rho_g)gd_p^3}{\mu^2}\right)^{0.213}$
Binary diffusivity	$D_{ji} = \frac{10^{-3}.T^{1.75}(1/M_i + 1/M_j)^{0.5}}{P_T(V_i^{0.33} + V_j^{0.33})^2}$
Diffusivity in mixture	$D_{jm} = \frac{(1-Y_j)}{\sum_{\substack{i=1 \\ i\neq j}}^{5}\left(\frac{Y_j}{D_{ji}}\right)}$

2. Model development

Of the many models proposed in the past, the two-phase model which considers the fluidized-bed reactor to consist of a bubble phase and a surrounding dense phase has proved to be the most suitable model, for incorporating recent findings on the hydrodynamics of fluidization (Werther, 1980). The hydrodynamic and transport property correlations are listed on Table 1. The details of the model is given elsewhere (Aljodai, 1999).

2.1. Assumptions

1. The bubble gas is devoid of solids and is in plug flow.
2. The extent of reaction in the bubble-cloud phase is negligible. This assumption is justified by the experimental evidence reported by Toor and Calderbank (1968) for small-size particles and high flow rate.
3. The dense phase is assumed to be perfectly mixed and uniform in temperature.
4. An average value of the bubble size and hence an average value of the interphase exchange parameter is used for the whole bed. The effective bubble size is taken as that which exists at 40% of the expanded bed height (Fryer & Potter, 1972). This assumption is widely used (Wagialla, Helal & Elnashaie, 1991; Elnashaie, Wagialla & Helal, 1991; Adris, 1989; Abshar, 1994).
5. The mass and heat transfer resistances between the particles and the dense-phase gas are negligible.

6. The ideal gas law applies to the gas phase in both phases.
7. The volumetric flow rate through the bubble phase is constant.

2.2. Reaction kinetics

Several kinetic studies have been reported in the literature for the oxychlorination route for 1,2-dichloroethane production (Carrubba, 1970; Fengqiu, Yongrong, Shunni & Gatang, 1994; Bakshi et al., 1991; Dmitrieva, Bakshi & Gelbshtein, 1991; Wachi & Asai, 1994).

In this work, kinetic study reported by Wachi and Asai (1994) for the process of 1,2-dichloroethane formation was used. This kinetic states that while the reaction exhibited first-order kinetics with regard to the concentration of cupric chloride, the dependency on ethylene concentration was interpreted by a Langmuir–Hinshelwood mechanism, however, the reaction sequence probably proceeds via chlorination of ethylene by cupric chloride. HCl and oxygen then regenerate the copper salt. The reaction rate has the following form:

$$r = \frac{k_r K_a C_E C_C}{1 + K_a C_E}$$

where

$$k_r = 269 \exp(-37.8/RT), \quad K_a = 0.63.$$

2.3. Mass and energy balances on bubble phase

The mass balance on component i and the energy balance equations in the bubble phase of the fluidized-bed reactor at steady state are given below:

Mass balances

$$\frac{dN_{ib}}{dh} = A_b(K_{bd})_i\left(\frac{N_{id}}{Q_d} - \frac{N_{ib}}{Q_b}\right),$$

at $h = 0$, $N_{ib} = N_{ibf}$ and $N_{id} = N_{idf}$.

Energy balance

$$Q_b \rho_g C_{pg} \frac{dT_b}{dh} = -A_b(H_{bd})_b(T_b - T_d),$$

at $h = 0$, $T_b = T_f$ and $T_d = T_f$.

2.4. Mass and energy balances on dense phase

The steady-state dense-phase mass and energy balances are given below:

Mass balance

$$N_{id} = N_{idf} + A_b U_b\left\{\left(\frac{N_{id}}{Q_d}\right) - \left(\frac{N_{ibf}}{Q_b}\right)\right\}[1 - e^{-\alpha H}] + V(1-\delta)(1-\varepsilon)r.$$

Energy balance

$$\rho_g C_{pg} Q_{df}(T_f - T_{ref}) - \rho_g C_{pg} Q_d(T_d - T_{ref}) + h_w A_w(T_w - T_d) + A_b U_b \rho_g C_{pg}(T_f - T_d)[1 - e^{-\beta h}] + V(1-\delta)(1-\varepsilon)(-\Delta H_r)r = 0,$$

where

$$\alpha = K_{bd}/U_b, \quad \beta = H_{bd}/U_b.\rho_g.C_{pg}.$$

3. Results and discussion

Sensitivity analysis was carried out to assess the various design, operating as well as hydrodynamic parameters on the reactor performance. The reactor design and operating conditions used in the simulations are listed in Table 2. The heat capacities, diffusivity, the density of the gas, and heat of reaction are functions of temperature.

3.1. Validation of model

The fluidized-bed model is tested against the pilot plant data reported by Cavaterra (1988) for a pilot plant working in Italy. It is clear from Table 3 that a very good agreement between the model predictions and industrial values have been obtained for temperature, C_2H_4 conversion and HCl conversion.

3.2. Effect of feed molar distribution

The ethylene molar flow rate was varied between 0.025 kg mol/s to 0.039 kg mol/s while HCl and oxygen (based on air) flow rates were kept at 0.064 and 0.0185 kg mol/s, respectively. Fig. 1 shows the HCl and oxygen conversion and also the EDC production rate. It is clear from Fig. 1 that as the ethylene molar flow rate is increased, the HCl and O_2 conversions and the EDC production increase. At low ethylene feed mole fractions (i.e., less than 0.1947) the ethylene conversion is high and this is because of the excess availability of HCl and oxygen and the shortage of ethylene quantity. After the ethylene feed ratio reaches 0.1947, the HCl and oxygen

Table 2
Design and operating data used in solving the reactor model

Design and operating data	Value
Bed height at min. fluidization	7.0 m
Bed diameter	3.4 m
Catalyst solid density	3075 kg/m^3
Catalyst particle density	1369 kg/m^3
Catalyst particle diameter	80 μm
Feed temp.	460 K
Cooling medium temp.	360 K

Table 3
Comparison between model predictions and pilot plant data at $HCl/C_2H_4 = 2$

Reactor Inlet	Plant	Model	
Res. Time, s	25	25	
Pressure, kPa	400	400	
HCl/C_2H_4(Feed)	2	2	
Reactor Outlet			Error%
Outlet Temp., K	498	498.18	+ 0.04%
C_2H_4 Conversion	99.3%	99.43%	+ 0.13%
HCl Conversion	98.3%	98.55%	+ 0.25%

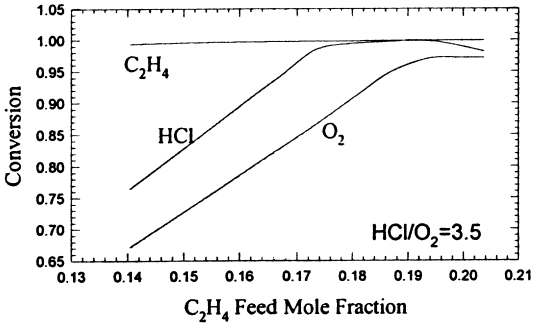

Fig. 1. Effect of C_2H_4 mole fraction on conversion.

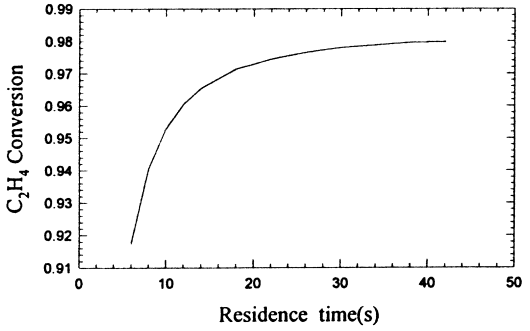

Fig. 2. The effect of residence time on C_2H_4 conversion.

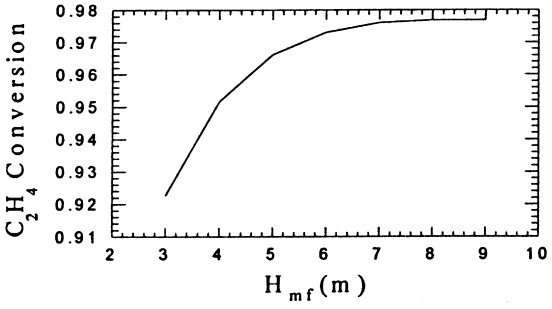

Fig. 3. Effect of height at minimum fluidization on C_2H_4 conversion.

conversions and EDC production rate remain constant while the ethylene conversion goes down because ethylene is no longer the limiting reactant.

3.3. Effect of residence time

Fig. 2 shows the effect of residence time on the ethylene conversion. As the residence time increases, the superficial velocity and the bubble size decrease and consequently C_2H_4 conversion increases. It is clear from Fig. 2 that the ethylene conversion increases asymptotically until it reaches a residence time of 20 s with ethylene conversion of 0.972, then the conversion increases slowly till the residence time of 40 s. At residence times in excess of 40 s, the conversion approaches an equilibrium conversion of about 0.98.

3.4. Effect of height at minimum fluidization

The bed height at minimum fluidization was varied between 3 and 9 m at a constant superficial velocity and bubble diameter. The increase of height at minimum fluidization at a constant velocity will increase the residence time because an increased in bed height leads to an increase in catalyst volume. The bubble diameter was kept constant at an average value which can be conducted through a proper distributor choice because a large bubble diameter has a negative effect on the ethylene conversion, since it decreases the interfacial area of mass and heat transfer between the bubble and dense phases.

Fig. 3 shows the changes in the height at minimum fluidization and the corresponding changes in the ethylene conversion. The figure also shows that small change of height at minimum fluidization has a significant effect on ethylene conversion until the height reaches 6 m. However, above 6 m height, there is no economic justification for the extra capital investments on reactor volume and catalyst quantity.

3.5. Effect of bubble diameter

The bubble size has a significant effect on the ethylene conversion. When the average bubble diameter (d_B) is 8 cm, an increase of bubble diameter by 50% will decrease the ethylene conversion from 96.5 to 86.5 as shown in Fig. 4. When the bubble diameter increases, the overall mass transfer area per unit bubble volume between bubble and dense phase decreases.

It is very important to take care of the bubble size by choosing a proper design for the reactant distributor in order to generate small bubbles. Furthermore, the reactor must be operated at sufficiently low superficial gas velocity. The growth of bubbles comes from their coalescence by one bubble catching up with another. Therefore, the bubbles coalescence may be reduced by fitting baffles inside the reactor to break up the bubbles and reduce their sizes.

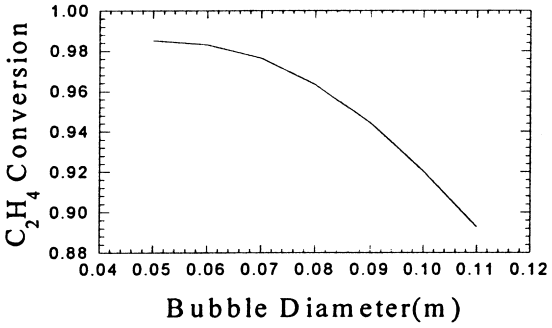

Fig. 4. Effect of bubble diameter on C_2H_4 conversion.

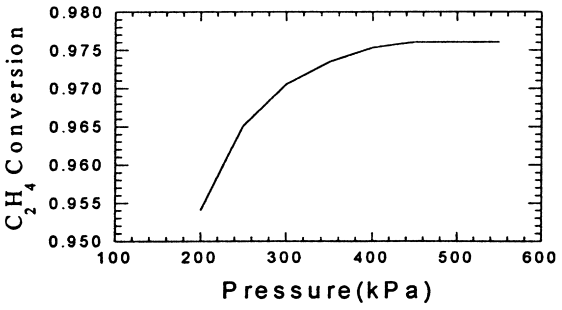

Fig. 5. Effect of pressure on C_2H_4 conversion.

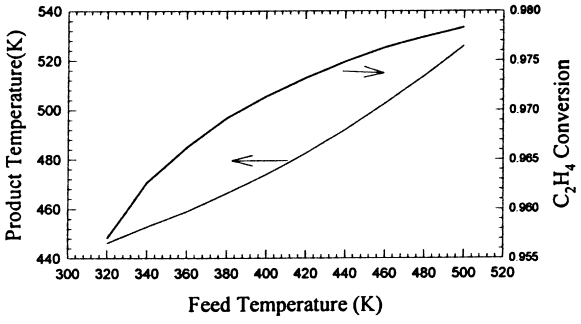

Fig. 6. Effect of feed temperature on C_2H_4 conversion and product temperature.

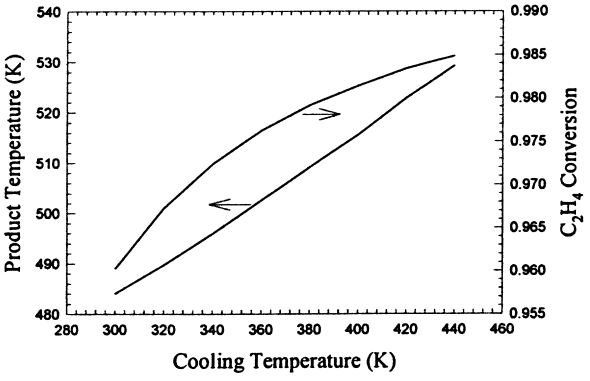

Fig. 7. Effect of cooling medium temperature on C_2H_4 conversion and product temperature.

3.6. Effect of total pressure

Increasing the total pressure from 200 to 400 kPa leads to an increase of ethylene conversion from 95.4 to 97.57%. Further increase of the total pressure above 400 kPa has a slight effect on the ethylene conversion and consequently EDC yield as shown in Fig. 5.

Pressure has a major control on density of gas. The density of gas is important since it affects the minimum fluidization, also it affects the bed voidage at minimum fluidization. Pressure has also an effect on the rate of reaction, since the pressure is a variable parameter of ethylene concentration and the increase of pressure will increase the ethylene concentration, therefore, it will increase the rate of reaction.

3.7. Effect of feed temperature on reactor performance

Fig. 6 shows the effect of variation of feed temperature on product temperature and ethylene conversion. The effect of feed temperature has a little effect on conversion, but has a large effect on product temperature. The feed temperature also affects the diffusivities of components in the gas mixture as shown. The diffusivity of each component in the gas mixture increase as the feed temperature increase. Furthermore, the diffusivities of the gas component affect the mass transfer coefficient. Each mass transfer coefficient increases as the diffusivity increases.

3.8. Effect of cooling medium temperature

Fig. 7 shows the effect of cooling medium temperature on ethylene conversion and product temperature. It is clear from Fig. 7 that when the cooling medium temperature increases from 300 to 440 K, C_2H_4 conversion increases from 0.955 to 0.985 while the product temperature increases from 482 to 528 K. The cooling medium temperature is a variable parameter of heat balance equation and any increase of cooling medium temperature will effect the heat removal term and the value of the product temperature.

4. Conclusions

A comprehensive reactor model that can be used to predict the performance of a fluidized-bed reactor for the ethylene oxychlorination to produce ethylene dichloride was developed. The model was verified by comparing simulation with industrial data obtained from the literature. The model predictions agree well with these industrial data. Ethylene and HCl conversions and reactor temperature were predicted with accuracy $\pm 2\%$ in a wide range of reaction conditions.

The results reveal that the ethylene feed composition has a very profound effect on the system performance. The parametric sensitivity analysis has shown that the system performance is particularly sensitive to residence time, bubble diameter and height at minimum fluidization.

The optimum ethylene conversion (i.e., 98%) and EDC yield of more than 98% at 98% HCl conversion can be achieved by working at a residence time of 25 s, height at minimum fluidization of 7 m and bubble diameter of 4.5 cm.

Notations

A_b	Cross sectional area of the bubble phase, m^2
d_B	Bubble diameter, m
C_C	Concentration of cupric chloride in the catalyst
C_E	Concentration of ethylene in gas phase, mol/m^3
C_{pg}	Molar heat capacity of gas, kJ/kg mol K
C_{ps}	Molar heat capacity of solid, kJ/kg mol K
h	Distance along bed height, m
H	Total bed height, m
H_{mf}	Height at minimum fluidization, m
h_w	Bed to cooling surface heat transfer coefficient, kJ/m^2 s K
H_{bd}	Interphase heat transfer coefficient between bubble and dense phase based on bubble phase volume, kJ/m^3 s K
K_{bd}	Interphase mass transfer coefficient between bubble and dense phase based on bubble phase volume, 1/s
k_r	Chemical reaction rate constant, 1/s
K_a	Adsorption equilibrium constant, m^3/mol
N_{ib}	Molar flow rate of component i in bubble phase, kg mol/s
N_{ibf}	Inlet molar flow rate of component j in bubble phase, kg mol/s
N_{id}	Molar flow rate of component i in dense phase, kg mol/s
N_{idf}	Inlet molar flow rate of component i in dense phase, kg mol/s
Q_b	Volumetric flow in bubble phase, m^3/s
Q_d	Volumetric flow in dense phase, m^3/s
r	Rate for reaction, kg mol/s m^3
R	Gas constant, kJ/kg mol K
T_b	Bubble phase temperature, K
T_d	Dense phase temperature, K
T_f	Feed gas temperature, K
T_w	Cooling medium temperature, K
ΔH_r	Heat of reaction, kJ/kg mol
U_b	Bubble velocity, m/s
U_{mf}	Minimum fluidization velocity, m/s

Greek letters

δ	Fraction of bed consisting of bubbles
ε	Void fraction
ρ_g	Density of the gas, kg/m^3
ρ_s	Density of solids, kg/m^3

References

Abshar, M. E. (1994). *Bifurcation, Stability and Chaos in fluidized bed catalytic reactors*. Ph.D. thesis, Univ. of Salford, Salford, UK.

Adris, A. M. (1989). *Steam reforming of methane*, MS. thesis., Univ. of Salford, Salford, UK.

Aljodai, A. (1999). *Modelling and bifurcation of 1,2 dichloroethane production by ethylene oxychlorination in fluidized bed reactor*. MS. thesis. King Saud University, Saudi Arabia.

Bakshi, Y. M., Gelbshtein, A. I., Gelperin, E. I., Dmitrieva, M. P., Zyshin, A. G., & Snagovshii, Y. S. (1991). *Study of the mechanism and kinetics of additive oxychlorination of olefins'. V. Kinetic model of the reaction*. Translated from *Kinetica Kataliz*, *32*, 663–672.

Carrubba, R. V. (1970). Kinetics of the oxychlorination of ethylene. *Industrial and Engineering Chemistry, Process Design and Development*, *9*, 414–419.

Cavaterra, E. (1988). Catalysts to make Dichloroethane. *Hydrocarbon Processing*, *67*, 63–67.

Dmitrieva, M. P., Bakshi, Y. M., & Gelbshtein, A. I. (1991). Investigation of mechanism and kinetics of additive oxychlorination of olefins. IV. Influence of copper containing salt catalyst with γ-Al_2O_3. Support on pinetic parameters of individual stages of reaction. *Translated from Kneticka Kataliz*, *32*, 74–80.

Elnashaie, S. S., Wagialla, K. M., & Helal, A. M. (1991). The use of mathematical and computer models to explore the applicability of fluidized bed technology for highly exothermic catalytic reactions. II. Oxidative dehydrogenation of ethyl benzene to styrene. *Mathematical and Computer Modeling*, *15*, 43–54.

Fengqiu, C., Yongrong, Y., Shunni, R., & Gatang, C. (1994). Studies on ethylene oxychlorination, II. Reaction mechanism and kinetics. *Petrochemical Technology*, *23*, 421–425.

Fryer, E., & Potter, Q. E. (1972). Bubble size variation in two-phase models of fluidized bed reactors. *Powder technology*, *6*, 312–322.

Keane, D. P., Stobaugh, R. B., & Townsend, P. L. (1973). Vinyl chloride. *Hydrocarbon Processing*, *52*, 99–104.

Markeloff, R. G. (1984). Convert oxychlorination from air to oxygen. *Hydrocarbon Processing*, *63*, 91–94.

Nawroski, J. S., & Velez, E. S. (1983). Oxychlorination of ethylene. *Applied Industrial Catalysis*, *1* (pp. 239–273). New York: Academic Press.

Reich, P. (1976). Air or oxygen for VCM. *Hydrocarbon Processing*, *55*, 85–89.

Smallwood, P. V., Stephenson, M. W., Newman, M. W., & Bunten, I. J. (1987). *Vinyl chloride polymers* (pp. 241–262). New York: Wiley.

SRI Report. (1998). Process Economic Program, Stanford Research Institute, Menlo Park, CA. *Ethylene and derivatives*, Vol. 1.

Toor, F. D., & Calderbank, P. H. (1968). *Proceedings of the tripartite chemical engineering conference*. Montreal, Que.: London Institution of Chemical Engineers.

Ullmann's Encyclopedia of Industrial Chemistry, (1986). Vol. 6, (5th ed.). (pp. 263–271). New York: VCH.

Wachi, W., & Asai, Y. (1994). Kinetics of 1,2 Dichloroethane formation from ethylene and cupric chloride. *Industrial Engineering and Chemistry Research*, *33*, 259–264.

Wagialla, K. M., Helal, A. M., & Elnashaie, S. S. (1991). The use of mathematical and computer models to explore the applicability of fluidized bed technology for highly exothermic catalytic reactions. I. Oxidative dehydrogenation of butene. *Mathematical and Computer Modeling*, *15*, 17–31.

Werther, J. (1980). Mathematical Modelling of fluidized bed reactors. *International Chemical Engineering*, *20*, 4.

Wimer, W. E., & Feathers, R. E. (1976). Oxygen gives low cost VCM. *Hydrocarbon Processing*, *55*, 81–84.