

# Method to Determine Molecular Diffusivities of Phenol in Bitumen

## Joseph S. Tang<sup>\*†</sup> and Pei Xin Zhang

China Institute of Atomic Energy, P.O. Box 275(104), Beijing 102413, China

anada has an estimated 400 billion m<sup>3</sup> of heavy oil deposit mainly located in Alberta and Saskatchewan. Heavy oil is a viscous tar-like liquid or semi-solid. It has a density of close to 1 and a viscosity as high as 1 million mPa·s at reservoir conditions. Due to its high viscosity, heavy oil cannot be produced by conventional means. A thermal process including cyclic steam stimulation, steam drive and Steam Assisted Gravity Drainage (SAGD) is by far the most cost-effective method to reduce viscosity, thereby producing heavy oil.

It has been reported that bitumen as well as conventional oil contains water soluble toxic compounds such as phenols (Tang, 1999; Speight, 1991), carboxylic acids, anhydrides, ketones and other high molecular weight acidic compounds generically classified as humic acids (Blum et al., 1986). During a heavy oil spill such as the one reported by Imperial Oil at Cold Lake in 1988 (Fitzhenry et al., 1992; Tang, 1999), the spilled bitumen can be in contact with the flowing water in the surface aquifer. These compounds, which are soluble both in oil and water, can leach slowly into the water contacted by the bitumen, posing a threat of groundwater contamination to the environment. To assess the environmental impact of groundwater contamination that could result from a spill, experiments were designed and conducted in the lab to study the rate of the release of phenol from bitumen into fresh flowing water.

The methods for measuring diffusion coefficients had been reviewed by Crank (1956). The methods are basically of two types according to whether the flow is steady or transient. In the steady state methods, diffusion coefficients can be calculated directly from the steady flow across a membrane or sintered disk. For the unsteady state transient experiments, the solute concentration distribution is determined at various times by optical means such as absorbance or refractive index, from which the diffusion coefficient can be deduced by comparing the data with a theoretical curve generated from a diffusion model. Alternatively, the diffusion coefficient can be measured by analyzing the cumulative flux leaving the diffusion medium into a well-stirred extraction phase as a function of time, a method known as the desorption method by Crank. However, the desorption method monitors the total weight loss in the transport phase, i.e., the cumulative flux, but not the concentration change in the leaching phase. Carman et al. (1954) suggested the calculation of diffusional flux and, hence, diffusion coefficient indirectly from the solute concentration in the extraction phase. This paper deals with the phenol transfer from a quiescent bitumen

\*Author to whom correspondence may be addressed. E-mail address: jtang@ cadvision.com

Phenols, carboxylic acids and other water-soluble compounds in bitumen constitute a potential source of groundwater pollution. To assess the extent of damages that could result from a spill, a water extraction method was used to determine the molecular diffusion of phenol in bitumen. This method involves the diffusion of phenol through bitumen into the flowing water in a well-stirred cell. From the phenol concentration in water, the phenol flux and hence the phenol diffusion coefficient in bitumen can be deduced. An approximate dimensionless solution was obtained by de-coupling the diffusional flux from the concentration buildup in water. The phenol diffusion coefficient was determined to be  $4.0 \pm 0.3 \times 10^{-8}$  m<sup>2</sup>/s at 22°C. The derivation and the properties of the dimensionless solution were also discussed in the paper.

Les phénols, les acides carboxyliques et autres composés solubles dans l'eau présents dans le bitume constituent une source potentielle de pollution des eaux souterraines. Afin d'évaluer l'étendue des dommages qui pourraient résulter d'un déversement. on a utilisé une méthode d'extraction de l'eau visant à déterminer la diffusion moléculaire du phénol dans le bitume. Cette méthode implique la diffusion de phénol dans le bitume présent dans l'eau en écoulement dans une cuve bien agitée. À partir de la concentration de phénol dans l'eau, on peut déduire le flux de phénol et donc le coefficient de diffusion du phénol dans le bitume. Une solution sans dimension approximative a été obtenue par découplage du flux diffusionnel de l'accumulation de la concentration dans l'eau. Le coefficient de diffusion du phénol a été déterminé à 4,0  $\pm$  0,3  $\times$  10<sup>-8</sup> cm<sup>2</sup>/s à 22°C. Le développement du calcul et les propriétés de la solution sans dimension sont également examinés dans cet article.

Keywords: groundwater contamination, 2-phase pollutant transfer, phenol diffusion coefficient in bitumen.

phase into the flowing water in a well-stirred cell. Upon making the approximation that the phenol transfer in bitumen and water can be de-coupled, the phenol concentration in water can be solved analytically.

<sup>†</sup>Present Address: JT Petrleum Consulting, 112 Edenwold Cr NW, Calgary, AB, Canada T3A 3T3

Among the few bitumen diffusion experiments reported in the literature, optical methods were used by Oballa and Butler (1989) for measuring the diffusion coefficient of toluene in bitumen and by Schmidt et al. (1982) for  $CO_2$  and other hydrocarbon gases in bitumen. For a trace amount of solute in bitumen, the optical method is not applicable, as the in-situ solute concentration in bitumen is too low to be measured by optical means. Accordingly, cumulative flux methods were employed by Tang (1999) for the study of phenol diffusion in water-in-bitumen emulsion and by Fu and Phillips (1979) for hydrocarbons in bitumen. Differences in methodology, experimental conditions and bitumen properties account for the inconsistent values reported in the literature.

## Theory

## **Closed System**

For a dilute solution where D and bitumen volume can be assumed to be constant, the molecular diffusion of phenol in bitumen can be modelled by the following second-order differential equation (Equation 1).

$$D\left(\frac{\partial^2 C}{\partial x^2}\right) = \frac{\partial C}{\partial t} \tag{1}$$

For a closed well-stirred cell (i.e., water flow rate q = 0 in Figure 1) and negligible mass transfer resistance across the interface, Equations (2a) and (2b) can be written to describe the phenol transport at the bitumen–water interface. The boundary conditions are:

a) At x = 0 and t > 0

$$DA\left(\frac{\partial C}{\partial x}\right) = V_{w}\left(\frac{\partial C_{w}}{\partial t}\right)$$
(2a)

$$\frac{C}{C_w} = K \tag{2b}$$

b) At x = L

$$\frac{\partial C}{\partial x} = 0 \tag{3}$$



Figure 1. Schematic of diffusion cell.

c) At 
$$t = 0$$

 $C = C_o$  for  $x \ge 0$  (in bitumen) and  $C_w = 0$ , for x < 0 (in water) (4)

As shown in boundary condition a), the diffusive phenol transport in bitumen is strongly coupled with the buildup of phenol concentration in the leaching water. An analytical solution has been given by Crank in the form of an infinite series for a slightly different boundary condition. However, the solution can be greatly simplified for slow molecular diffusion in bitumen at an early time when only a negligible amount of phenol is leached into the water, i.e.,  $L^2/Dt >> 10$  so that the semi-infinite boundary condition can be applied. Furthermore, under the slow-transfer, small-flux condition, phenol transport in bitumen can be practically de-coupled from the phenol concentration in the leaching water by approximating zero phenol concentration at the interface. This approximation is valid for slow transfer at early time with large volume of extraction water so that the phenol concentration in water is much lower than the initial phenol concentration in bitumen, i.e.,  $C_0 >> C_w$ . Then Equation (2b) becomes:

At 
$$x = 0$$
  $t > 0$   $C = 0$ 

and the standard semi-infinite solution can be applied.

$$\frac{C(x,t)}{C_0} = erf\left(\frac{x}{2\sqrt{Dt}}\right)$$
(5)

where the error function  $erf(\xi)$  is defined as:

$$erf(\xi) = \frac{2}{\sqrt{\pi}} \int_0^{\xi} \exp(-\xi^2) d\xi$$
(6)

The phenol flux into the water phase, / (mg/s), can be evaluated from Equations (2a) and (5):

$$J = DA \left(\frac{\partial C}{\partial x}\right)_{x=0} = C_o A \sqrt{\frac{D}{\pi t}}$$
(7)

By integrating Equation (7), the water-phase phenol concentration can be derived:

$$C_{w}(t) = \frac{1}{V_{w}} \int_{0}^{t} \int dt = \frac{2C_{o}A}{V_{w}} \sqrt{\frac{Dt}{\pi}}$$
(8)

This equation was used by Tang (1999) to determine the phenol diffusion coefficient in water-in-bitumen emulsion. An equivalent form known as the initial rate method was discussed by Crank.

#### **Open System**

For an open system as shown in Figure 1 with q > 0, phenol concentration in water can be expressed by the following mass balance equation.

$$\frac{dC_w(t)}{dt} = \frac{J(t) - qC_w(t)}{V_w}$$
(9)

The flux J(t) is given by Equation (7). Combining with the decoupled initial condition  $C_w(t) = 0$  at t = 0, Equation (9) can be solved by Laplace transform.

$$C_{w}(t) = 2 \frac{C_{o}A}{V_{w}} \sqrt{\frac{DV_{w}}{q\pi}} \exp\left(-\frac{qt}{V_{w}}\right) \int_{0}^{\sqrt{\frac{qt}{V_{w}}}} \exp(\lambda^{2}) d\lambda \qquad t \ge 0 \quad (10)$$

The integral can be evaluated by numerical integration.

#### **Dimensionless Solution**

Equation (10) can be expressed in a dimensionless form:

$$C_{wD}(t_D) = \exp(-t_D) \int_0^{\sqrt{t_D}} \exp(\lambda^2) d\lambda$$
 (11)

where the dimensionless concentration and time are defined as:

$$C_{wD}(t_D) \equiv \frac{C_w(t)}{2\frac{C_oA}{V_w}\sqrt{\frac{DV_w}{q\pi}}} \qquad t_D \equiv \left(\frac{q}{V_w}\right)t \qquad (12)$$

The dimensionless solution is shown graphically in Figure 2.

#### Limiting Solution

A limiting solution at large  $t_D$  can be obtained by applying l'Hopital rule to Equation (11):

$$\lim_{t_D \to \infty} C_{wD}(t_D) = \frac{1}{2\sqrt{t_D}}$$
(13)

Similarly, it can be demonstrated that a limiting slope of 1

exists for the  $C_{wD}$  vs  $\sqrt{t_D}$  plot.

**Peak Time, Peak Concentration and Normalized Solution** The dimensionless peak time  $t_{D,max}$  can be evaluated by setting  $dC_{D,w}(t_D)/dt_D$  (Equation 11) equal to zero:

$$\int_{0}^{\sqrt{t_{D,\max}}} \exp(\lambda^2) d\lambda = \frac{\exp(t_{D,\max})}{2\sqrt{t_{D,\max}}}$$
(14)

Solving Equation (14) numerically for  $t_{D,max}$ , we have:

$$t_{D,max} = 0.854$$
 (15a)

and the peak concentration  $C_{wD,max}$  ( $t_{D,max}$ ) is:

$$C_{wD,max} = C_{wD}(t_{D,max}) = 0.541$$
 (15b)

Normalizing Equation (10) by the peak time and peak concentration (Equations 15a and 15b) results in the following normalized solution Equation (16):

$$C_{wN}(t_N) = 1.848 \exp(-0.854t_N) \int_0^{0.924\sqrt{t_N}} \exp(\lambda^2) d\lambda$$
 (16)



Figure 2. Dimensionless concentration vs. time plot.

The normalized concentration  $C_{wN}(t_N) = C_w(t)/C_{w,max}$  and normalized time  $t_N = t/t_{max}$ .

## Experimental Procedures

Bitumen produced from a field in Northern China was used for the tests. A 19 cm diameter glass crystallizing dish fitted with an inlet and outlet port was used for the diffusion test (Figures 1). The inlet port was connected to a constant rate 2-head Beckman pump and the effluent from the outlet port was collected at constant intervals with a modified Gilson automated fraction collector. 600 g of distilled water was placed into the dish, and a magnetic bar was used to facilitate a mild agitation of the water to create a uniform phenol concentration in the water phase. Since the densities of the bitumen and distilled water were about equal at 22°C, a screen was necessary to support the bitumen so as to provide a stable bitumen-water interface. A stainless steel 40 mesh (425 µm) screen was cut and inserted into the dish at slightly below the water level. 1480 g (1500 mL) of bitumen was transferred carefully onto the top of the screen. The top of the dish was covered with a glass plate, which was then sealed with a silicone sealant. Distilled water was pumped into the cell at 240 mL/h and 10 mL of effluent sample was collected hourly for phenol analysis. The experiment was stopped after 30 days. To avoid phenol degradation by bacteria, the samples were immediately preserved with strong sulfuric acid at pH 1 prior to phenol analyses. A standard non-specific colorimetric method (Nagudat code 6537L) was adopted for all routine water-phase phenol analyses (Tang 1999). This method has a detection limit in the 5 to 10 ppb range. The original phenol content in the bitumen was determined by hot water extraction.

## **Results and Discussion**

Listed in Table 1 are the experimental parameters for the diffusion test. After correcting for the holdup of 4.5 mL from the production volumes, two methods were used to estimate the diffusion coefficient.

## **Method 1: Asymptotic Limiting Solution**

As indicated in the limiting solution Equation (13), the dimensionless concentration  $C_{wD}$  should vary linearly with at late time with a slope approaching 0.5. The dimensionless slope can be related to the physical slope through Equation (12) as:

Temperature, (°C)	22
Diffusion cell dimension	
Diameter, (cm)	19.0
Cross-section area A, (cm <sup>2</sup> )	283.5
Height, (cm)	10.0
Holdup volume, (mL)	4.5
Bitumen	
Volume, (mL)	1500*
Thickness L, (cm)	5.29
Viscosity @ 22°C, Pa-s	48.5
@ Shear rate <10 s <sup>-1</sup>	
Density @ 22°C, (g/mL)	0.984
Phenol Content $C_{\alpha}$ (mg/L)	4.50**
Distilled water	
Volume V <sub>w</sub> (mL)	600*
Water flow rate $q$ , (mL/hr)	240
Sampling interval. (h)	1

Physical limiting Slope = 
$$\lim_{t \to \infty} \left[ \frac{dC_w}{dt^{-1/2}} \right] = \frac{AC_o}{q} \sqrt{\frac{D}{\pi}}$$
(17)

Therefore, the diffusion coefficient can be obtained from the slope of the limiting asymptote in the  $C_w$  vs  $1\sqrt{t}$  plot. However, the asymptote may be difficult to construct from the actual data due to the experiment not being carried out far enough into the late time region. As well, the accuracy may suffer as only the data points in the late time region are utilized in the calculation. As an improvement to the method, the diffusion coefficient obtained from the limiting slope (Equation 17) is used as an initial guess and the value of diffusion coefficient is adjusted until Equation (10) gives the best fit of all the data over the entire range.

The limiting slope was estimated from the experimental data in Figure 3. A straight line passing through the origin was drawn through the data points at late time and the limiting slope was determined to be 0.182 (day<sup>0.5</sup> mg/L). Based on Equation (17) and the values of A,  $C_o$  and q listed in Table 1, the limiting diffusion coefficient D was calculated to be  $4.2 \times 10^{-8}$ cm<sup>2</sup>/s. This estimate served as an initial guess for the fitting process. Upon trial and error, the best fit was found to be D = $4.0 \times 10^{-8}$  cm<sup>2</sup>/s. The best fit and the sensitivity of the match are shown in Figure 4 at 3 different diffusion coefficient values, from which the phenol diffusion coefficient in bitumen was determined to be  $4.0\pm0.3 \times 10^{-8}$  cm<sup>2</sup>/s. The theoretical curve for  $D = 4.0 \times 10^{-8}$  cm<sup>2</sup>/s (best fit) is plotted in Figure 3 along with the data points for comparison.

#### Method 2: Peak Method

Diffusion coefficient and peak concentration are related through Equations (15b) and (12). Because of the data noise, it may be difficult to locate the concentration peak and its concentration. However, peak time depends only on the ratio of  $q/V_w$  and is independent of the diffusion coefficient. Phenol concentration peaks at about  $t = 0.854 * V_w/q$ , which is 2.1 d for the test. Similar to the limiting asymptote method, the estimated peak concentration is used as a starting point in the simulation.







Normalized solution Equation (16) was utilized for matching the test data. Various values of  $C_{w,max}$  were tried to normalize the phenol concentration and the normalized data were compared with the theoretical normalized curve Equation (16) in Figure 5. Not surprisingly, the best fit was at  $C_{w,max} = 0.121$ mg/L which corresponds to a diffusion coefficient of  $4.0 \times 10^{-8}$ cm<sup>2</sup>/s from Equation (15). The sensitivity of the fit is also shown in Figure 5. The value  $4.0(0.3 \times 10^{-8} \text{ cm}^2/\text{s}$  thus determined is identical to that for method 1 because of the complementary nature of these two methods. The good match of data for both methods validates the solution and hence the de-coupling approximation.

#### Phenol Distribution in Bitumen

Equation (5) was used to calculate the phenol distribution in bitumen at t = 50 days at the best-fit value of  $D = 4.0 \times 10^{-8}$  cm<sup>2</sup>/s. The calculation revealed that even at a contact time of 50 d, the entire phenol concentration gradient was still within a zone 1.5 cm from the interface, and the phenol did not sense the presence of the boundary located at L = 5.29 cm. This is consistent with the semi-infinite assumption made in the previous derivation. Moreover, there were only 7% of the total phenol transferred to the fresh water in 30 d based on the mass



Figure 5. Normalized phenol concentration vs. normalized time at various diffusion coefficients.

balance calculation. This small cumulative flux justified the decoupling approximation.

Tang (1999) documented the measurement of phenol diffusion coefficient in water-in-bitumen emulsion using the water extraction method in a closed system. In his method, water was added to replenish the volume loss due to the samples taken out for phenol analysis. The coefficient was found to be 2.23 imes10<sup>-8</sup> cm<sup>2</sup>/s at 4°C which is in good agreement with the value  $4.0 \times 10^{-8}$  cm<sup>2</sup>/s at 22°C reported in this paper. The open system described in this paper has the merit over the closed system that the water in the leaching cell is continuously refreshed with water influx, there would be less accumulation of soluble materials from the bitumen which could affect the phenol solubility in the leaching water. It will also reduce biological and chemical degradation of phenol that may occur during the test. Another drawback of the closed system is that continuous buildup of phenol concentration in water may invalidate the assumption of de-coupling at late time. In comparison with other data published in the literature, our value agrees in order of magnitude with Fu and Phillip's values of  $7.8 \times 10^{-8}$ cm<sup>2</sup>/s for toluene in bitumen and  $8.2 \times 10^{-8}$  cm<sup>2</sup>/s for benzene in bitumen at 23°C. It is lower than the value  $50 \times 10^{-8}$  cm<sup>2</sup>/s at infinite dilution by Oballa and Butler (1989) for toluene in bitumen at 22°C. The high value of Oballa and Butler may be attributed to the inaccurate measurement of the in-situ toluene concentration distribution in an opaque system and different bitumen used for the tests.

## Conclusions

- In the study of phenol transport from viscous bitumen into flowing water in an open, well-stirred cell, an approximate solution was obtained by de-coupling the phenol flux from the buildup of phenol concentration in water. A good fit of data validated the de-coupling approximation.
- 2. The dimensionless solution predicted a dimensionless peak concentration of 0.541, a dimensionless peak time of 0.854 and the existence of a limiting solution at late time where the dimensionless concentration varied linearly with the reciprocal of the square root of the dimensionless time at a slope of 0.5. The peak concentration and the late time

solution are useful in providing an estimate of phenol diffusion coefficient to be used as an initial guess for subsequent fine matching of the test data.

- 3. The phenol diffusion coefficient was obtained by matching the entire range of test data with the dimensionless solution. The diffusion coefficient of phenol in bitumen was determined to be  $4.0 \pm 0.3 \times 10^{-8}$  cm<sup>2</sup>/s at 22°C.
- 4. The phenol concentration gradient in bitumen was limited to within a zone of 1.5 cm from the bitumen-water interface at the end of 30 d, during which only 7% of the phenol was extracted. The small flux justified the approximations of semi-infinite medium and transport de-coupling.
- Water leaching is a valid method to determine the diffusion coefficients of trace, water-soluble compounds in bitumen. Optical method is not applicable due to low solute concentration.
- The de-coupled solution is also useful in estimating the extent of groundwater contamination in a flowing aquifer resulting from a bitumen spill.

## Acknowledgements

The authors wish to thank the China Institute of Atomic Energy and the China Environmental Protection Agency for permission to publish this work.

## Nomenclature

Α	interface area, (cm <sup>2</sup> )
С	phenol concentration in bitumen, (mg/mL)
C <sub>o</sub>	initial phenol concentration in bitumen, (mg/mL)
C <sub>w</sub>	phenol concentration in leaching water, (mg/mL)
C <sub>wD</sub>	dimensionless phenol concentration
C <sub>wD.max</sub>	dimensionless phenol peak concentration
C <sub>w.max</sub>	peak phenol concentration, (mg/mL)
C <sub>wN</sub>	normalized phenol concentration, (C/C <sub>w.max</sub> )
D	molecular diffusion coefficient of phenol in bitumen, (cm <sup>2</sup> /s)
Erf(ξ)	error function with argument ξ
J	phenol flux into leaching water, (mg/s)
Κ	partition coefficient
L	thickness of the bitumen layer, (cm)
q	water flow rate, (mL/s)
t	time, (d)
t <sub>D</sub>	dimensionless time, $(tq/V_w)$
t <sub>D,max</sub>	dimensionless phenol peak time
t <sub>max</sub>	phenol peak time, (s)
t <sub>N</sub>	normalized time, (t/t <sub>max</sub> )
Vw	volume of water, (mL)
x	distance from the interface, (cm)

## **Greek Symbols**

- $\lambda$  integration parameter
- $\xi$  argument for the error function

## References

- Blum, D.J.W., R. Hergenroeder, G.F. Parkin and R.E. Speece, "Anaerobic Treatment of Coal Conversion Wastewater Constituents: Biodegradability of Toxicity", J. Wat. Pol. Control Fed. **58**, 122–127 (1986).
- Carman, P.C., and R.A.W. Haul, Proc. Roy. Soc. 222 A, 109-112 (1954).
- Crank J., "The Mathematics of Diffusion", Oxford University Press Oxford, UK (1956), pp. 219–257.
- Fitzhenry, L.C., R.M. Bacon and J.M. Gronseth, "Reservoir/Well Repair and Environmental Impact Management of Cold Lake C7-13 Surface Discharge", J. Petrol. Technol., May (1992), pp. 618–622.

- Fu, B.C.H. and C.R. Phillips "New Technique for Determination of Diffusivities of Volatile Hydrocarbons in Semi-Solid Bitumen", Fuel 59, 557–560 (1979).
- Oballa, V. and R.M. Butler, "An Experimental Study of Diffusion in the Bitumen-Toluene System", J. Can. Petrol. Technol. 28, 63-69 (1989).
- Speight, J.G., "The Chemistry and Technology of Petroleum", 2nd ed., Marcel Dekker, New York, NY (1991), pp. 69–72.
- Schmidt, T., T.H. Leshchyshyn and V.R. Puttagunta, "Diffusivity of Carbon Dioxide in Athabasca Bitumen", Petrol. Soc. of CIM, 33rd Annual Tech Meeting in Calgary, CIM 82-33-100, June 6-8 (1982).
- Tang, J.S., "Experimental Study of Groundwater Contamination by Phenol from Water/Heavy Oil Emulsion", Paper 95–85, Special Edition Can.J.Petrol.Technol. **38**, 47, (1999).

Manuscript received August 6, 1999; revised manuscript received May 15, 2000; accepted for publication July 13, 2000.

## Chemical Engineering Research and Design

As part of a cooperative agreement between the Institution of Chemical Engineers (UK) and The Canadian Society for Chemical Engineering, the *table of contents* of each issue of the respective research journals will appear in the other. This journal is available to members of The Canadian Society for Chemical Engineering for \$131.00.

### Chemical Engineering Research and Design

Vol. 78, No. A8 November 2000

Special issue Separation Processes

## Contents

1059 Editorial

## **Special Papers**

- 1061 Absolute Determination of the Composition of Binary Gas Mixtures by Admixture of Known Components. *M.J. Heslop, G. Mason and B.A. Buffham*
- 1066 Air Separation by Polysulfone Hollow Fibre Membrane Permeators in Series: Experimental and Simulation Results. H.M.S. Lababidi
- 1077 Predicting Solvent Effects on Reactions and Liquid-Liquid Equilibrium by Computer Simulation. *M. Sheehan and P. Sharratt*
- 1084 Zeolite Membranes for Use in Alcohol/Water Separations Part I: Experimental Investigation. S.M. Holmes, M. Schmitt, C. Markert, R.J. Plaisted, J.O. Forrest,
- P.N. Sharratt, A.A. Garforth, C.S. Cundy and J. Dwyer
- 1089 Hybrid membrane Adsorbents for Fluidized Bed Separations. B. Bhardwaj, S.P. Perera and B.D. Crittenden

1098 Advances in Cyclone Modelling Using Unstructured Grids. M.D. Slack, R.O. Prasad, A. Bakker and F. Boysan

#### **General Papers**

- 1105 Blending of Newtonian and Shear-thinning Fluids in a Tank Stirred with a Helical Screw Agitator. J. Aubin, I. Naude, J. Bertrand and C. Xuereb
- 1115 Use of Neural Networks as a Tuning Method for an Adaptive PID: Application in a Heat Exchanger. *C. Riverol and V. Napolitano*
- 1120 A Comparison of Dynamic Models for an Evaporation Process.
  - N.T. Russell, H.H.C. Bakker and R.I. Chaplin
- 1129 On the Flow and Vortex Shedding Characteristics of an In-Line Tube Bundle in Steady and Pulsating Crossflow.
- E. Konstantinidis, D. Castiglia, S. Balabani and M. Yianneskis 1139 Gas Hold-Up in Aerated Slugging Columns
- M.N. Coelho Pinheiro, A.M.F.R. Pinto and J.B.L.M. Campos 1147 In-Process Particle Characterization by Spectral Extinction.
- S. Gordon, R. Hammond, K. Roberts, N. Savelli and D. Wilkinson
- 1153 An Experimental Investigation of Adiabatic Two-Phase Flow in a Vertical Pipe at Different Temperatures. *M.R. Malayeri, J.M. Smith and H. Müller-Steinhagen*
- 1160 List of Referees
- 1161 Index 2000