A NUMBER BALANCE APPROACH TO THE MODELLING OF THE LEACHING OF PARTICLES IN MULTIPLE WELL-MIXED TANKS

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A dissertation submitted to the Faculty of Engineering, University of the Witwatersrand, Johannesburg, in fulfilment of the requirements for the degree of Master of Science in Engineering

Johannesburg, 1985

DECLARATION

I declare that this dissertation is my own, unaided work. It is being submitted for the Degree of Master of Science in Engineering in the University of the Witwatersrand, Johannesburg. It has not been submitted before for any degree or examination in any other University.

P.H.COMNINOS

26th day of June 1985

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P.H.COMNINOS

28th day of June 1985

ABSTRACT

The aim of this research is to model the continuous leaching of particles in multiple well-mixed tanks, taking into consideration the particle size distribution of the feed. It is desired that the model answer such questions as the tank volumes required for a given leach efficiency.

The model is developed using a particle number balance over each tank in the system. It is assumed that each particle decreases in size uniformly according to the ideal shrinking particle model. Experiments consisting of the leaching of iron particles by a ferric sulphate solution are conducted to ascertain how the model can be applied to a practical system.

The major conclusions of the research are that a single tank can be successfully modelled using a dimensionless graphical technique, which allows one, for any feed particle size distribution, to determine the volume of the tank required for a given leach efficiency. For multiple stirred tanks in series the model is able to predict the conversion over each tank, but an iterative procedure is required to determine the volume of the set of tanks to yield a required overall leach efficiency.

ACKNOWLEDGEMENTS

I would like to thank Professor A.W. Bryson for his invaluable advice and assistance.

I would also like to thank MINTEK for their financial assistance, and the Chemical Engineering Department of the University of the Witwatersrand for extending the use of its facilities to me.

Για την Νικολεττα και την οικογενεια μου

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1 INTRODUCTION

The mineral industry is an integral part of South Africa's economy and new and improved processes to extract minerals from their ores are continuously being sought. Of these the hydrometallurgical methods are being preferred over the pyrometallurgical methods since they yield a better quality product, are less polluting and in many cases are more efficient.

The aim of this research is to derive a simplified model to describe leaching in multiple well-mixed tanks in series. The model will attempt to answer such questions as the volume and the number of tanks required to achieve a given leach efficiency as well as the leach efficiency obtained from a given number and volume of tanks. The model was developed from particle number balance over the i^{th} tank in a series of n well-mixed tanks. The balance gave a first order differential equation which was solved to give the size distribution and the ratio of the number of particles entering to those exiting the i^{th} tank and thus the conversion. To simplify the algebra of the model the chemical reaction of the particles was described by the shrinking particle mechanism (Levenspiel, 1972). The model can however be extended to incorporate other reaction systems.

Laboratory-scale experiments were conducted to verify the model and entailed the leaching of iron particles by a ferric sulphate solution in two well-mixed tanks placed in series. This reaction was chosen for its simple kinetics, but it must be borne in mind that any reaction whose kinetics are known can used in the model.

A literature survey was conducted but revealed few relevant articles. Those that were found are presented here. Levenspiel (1972) applies the residence time distribution of a well-mixed tank to the shrinking core and shrinking particle models to obtain the leach efficiency in a single tank for a feed of uniform particle size. He includes a numerically given feed particle size distribution by applying his model over each of the discrete size ranges. He does not predict the size distribution in the outlet of the tank and therefore his equations cannot be extended to multiple tanks.

Loveday (1975) bases his equations on a system involving a uniform size feed to a single tank with an overflow of leach liquor and with the complete dissolution of the particles in the tank. The size distribution in the tank is easily calculated since there will be an equal number of particles in each size range due to the complete dissolution of the particles in the tank. The model lends itself easily to the incorporation of a feed particle size distribution but is not extendable to include multiple well-mixed tanks.

Sepulveda and Herbst (1978) use the particle balance approach but do not consider expressing the leach efficiency in terms of the moments of the feed particle size distribution and a dimensionless number that is a function of the system variables which simplifies the model.

Treybal (1981) describes an approach that uses experimental data, but this requires a new set of experiments for each configuration of the system variables.

2 THEORY

The ideal shrinking particle model (Levenspiel, 1972) is a model to describe the way a particle behaves upon reacting. It has two basic assumptions, the first being that the particle is non-porous and therefore reaction occurs only at the surface of the particle. The second is that reacted material is released from the particle without affecting the reaction rate.

The ideal shrinking particle model can be expressed mathematically

$$\begin{aligned} dm(\ell)/dt &= -k_i\psi_i a(\ell)\omega \\ &= -k_i\psi_i\phi\ell^2\omega \\ &= 3\rho\ell^2 u_i\omega \end{aligned}$$

where: $a(\ell)$ = exposed surface area of particle of size ℓ

k_i = rate constant in tank i

ℓ = characteristic particle size

= $[m(\ell)/\rho]^{1/3}$

$m(\ell)$ = mass of single particle of size ℓ

t = residence time of particle in contact with solution

u_i = particle growth rate in tank i

= $d\ell/dt$

ϕ = shape factor

= $a(\ell)/\ell^2$

ψ = quantity dependant on chemical and physical properties of the solution environment in tank i

ρ = density of particle

ω = molecular weight of dissolving species

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ρ = density of particle

ω = molecular weight of dissolving species

Therefore

$$u_i = -k_i \psi_i \phi \omega / 3\rho \qquad \ldots(1)$$

ie. the ideal shrinking particle model assumes u_i is independent of ℓ.

Conducting a particle balance for the i^{th} tank, in the size range ℓ to $\ell+d\ell$, ie.

the number of particles entering the i^{th} (from tank i-1) = the number of particles leaving the i^{th} tank + the number of particles entering the size range ℓ to $\ell+d\ell$ as a result of reaction.

and assuming :

1. the system is at steady state.
2. the tanks are well mixed.
3. the ideal shrinking particle model applies.
4. the volume flowrate through the tanks is constant.

this can be mathematically expressed as: (See "Appendix A. Detailed theory" on page 35)

$$df_i(\ell)/d\ell - \alpha_i f_i(\ell) = -\beta_i \alpha_i f_{i-1}(\ell)$$

where: $f_i(\ell)d\ell$ = fraction of particles in tank i with sizes between ℓ and $\ell+d\ell$

N_i = no. of particles in tank i per unit volume of solution

Q = volumetric flowrate of solution

u_i = particle growth rate in tank i

$= d\ell/dt$

$V_i = $ volume of tank i

$\alpha_i = -Q/u_i V_i$

$\beta_i = N_{i-1}/N_i$

integrating this gives

$$f_i(\ell) = \alpha_i \beta_i \exp(\alpha_i \ell) \int_{\ell}^{\infty} \exp(-\alpha_i \ell') f_{i-1}(\ell') d\ell' \qquad \ldots(2)$$

where: $$1/\beta_i = 1 - \int_0^{\infty} \exp(-\alpha_i \ell') f_{i-1}(\ell') d\ell' \qquad \ldots(3)$$

and using this equation the following moments may be calculated

$$\eta_i = \int_0^{\infty} \ell \, f_i(\ell) d\ell = \beta_i \eta_{i-1} - 1/\alpha_i \qquad \ldots(4)$$

$$\sigma_i = \int_0^{\infty} \ell^2 f_i(\ell) d\ell = \beta_i \sigma_{i-1} - 2\eta_i/\alpha_i \qquad \ldots(5)$$

$$\xi_i = \int_0^{\infty} \ell^3 f_i(\ell) d\ell = \beta_i \xi_{i-1} - 3\sigma_i/\alpha_i \qquad \ldots(6)$$

Therefore ξ_i can be obtained in terms of η_0, σ_0 and ξ_0 and the overall fractional conversion can be expressed in terms of the third moment as shown in Appendix A.

$$C = 1 - (\xi_n/\xi_0) \prod_{i=1}^{n} 1/\beta_i \qquad \ldots(7)$$

Conversion over one tank

Using the above equations and defining:

$$t_i^* = -\eta_0/u_i$$

where t_i^* is the time taken for the average sized particle to be completely leached.

$$T_i = (V_i/Q)/t_i^*$$

the following can be derived by combining equations (1) through(7) (See "*Appendix A. Detailed theory*" *on page* 35)

$$C_i = [3(\sigma_0/\eta_0^2)T_i - 6T_i^2 + 6T_i^3/\beta_i](\eta_0^3)/\xi_0 \qquad \ldots(8)$$

where:

$$T_i = (\omega\phi/3\rho)(V_i/Q)k_i\psi_i/\eta_0 \qquad \ldots(9)$$

$$1/\beta_i = 1 - \int_0^\infty \exp(-\alpha_i \ell')f_0(\ell')d\ell' \qquad \ldots(10)$$

$$\alpha_i = 1/\eta_0 T_i \qquad \ldots(11)$$

The size distribution in the outlet stream is given by

$$f_i(\ell) = \alpha_i\beta_i\exp(\alpha_i\ell)\int_\ell^\infty \exp(-\alpha_i\ell')f_0(\ell')d\ell' \qquad \ldots(12)$$

Using equation (8) a graph of C vs. T can be plotted. Using the graph in conjunction with (9) the required tank volume for a particular conversion can be calculated noting that ψ_i is a function of the concentrations in the tank *and thus also of* the conversion.

A plot of C vs. T was made using the size distribution used for the experiments. The size distribution is calculated in Section 3.3 and the plot is shown in Figure 1 on page 7.

The detailed derivation for the above theory can be found in "Appendix A. Detailed theory" on page 35.

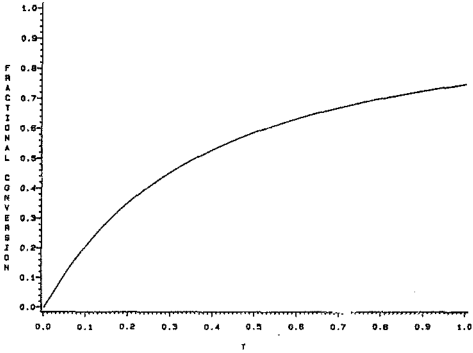

Figure 1. Plot of fractional leach efficiency vs T

3 EXPERIMENTAL METHOD

3.1 SELECTION OF THE REACTION

A reaction that *approximates* ideal shrinking particle behaviour was required. For this a particle satisfying the following conditions is needed. The particle needed to:

1. have few impurities.

2. not be too irregular.

3. form no ash layer upon leaching.

4. be of a manageable size.

An additional requirement for the reaction was that *it should be easy to* model.

It was decided that the leaching of iron particles by ferric sulphate would constitute a suitable reaction for the following reasons.

1. Iron particles were obtainable with a maximum of two percent impurities.

2. The particles were observed through a microscope and found to satisfy the above conditions. The photograph has been reduced, for convenience, from the original. (see "Appendix M. Photograph of the iron particles" on page 72)

3. Iron forms no ash layer upon leaching.

The ferric sulphate was not readily available so it was formed by oxidising ferrous sulphate using hydrogen peroxide.
The method is described in Section 3.2

3.2 OXIDATION OF FERROUS TO FERRIC SULPHATE

Ferrous sulphate is oxidised as follows.

$$2FeSO_4 + H_2SO_4 + H_2O_2 \rightarrow Fe_2(SO_4)_3 + 2H_2O$$

A low pH was desirable since at high pH's the ferric sulphate is insoluble. However the pH could not be too low since this causes a significant increase in the reaction of the hydrogen ions with the iron.

The solution storage capacity for each run was 35 litres and to produce this the following reactants are required.

35 ℓ	H_2O
7 kg	$FeSO_4.7H_2O$
680 mℓ	98% H_2SO_4
1 ℓ	30% H_2O_2

(see "Appendix C. Oxidation of ferrous sulphate" on page 44)

The solution was analysed as follows:

1. the total aqueous iron concentration was obtained by A.A. spectroscopy.
2. the ferric ion concentration was determined by the titration as shown in "Appendix D. Titration for Ferric ion" on page 46.
3. the ferrous ion concentration was determined from the difference of the above two concentrations.
4. the H_2SO_4 concentration was determined by the titration as shown in "Appendix E. Titration for Sulphuric acid" on page 47.

3.3 FEED PARTICLE SIZE DISTRIBUTION

The *number density* function of the particles was obtained from two independent methods.

For the first method photographs were taken through a microscope of the particles on millimetre graph paper (see "Appendix M. Photograph of the iron particles" on page 72). It was seen that the particles approximated ellipsoids and the maximum and minimum diameters of each particle measured. These measurements were used in a program (see "Appendix G. Program 1" on page 49) to calculate the size distribution and shape factor. The shape factor ϕ was calculated by the program to be 4,7 .

A KONTRON IBAS image analyser was used for the second method. The image analyser uses a T.V camera to *scan a mounted polished sample and* then calculates the distribution using statistical methods (King, 1984). The outputs from the analyser are in the form graphs of the particle size vs. the mass fraction smaller than that size. Four runs were made and The graphs are shown in "Appendix L. Graphs from the image analyser" on page 70. A program (see "Appendix H. Program 2" on page 52) was used to evaluate the number density function from these graphs.
The moments were calculated and the first moment, i.e. the mean particle size, was found to be 200 μm.

Particles less than 100 μm were not considered since they constituted a very small fraction of the mass and were unreliable due to noise in the graphs which is amplified upon conversion from mass fraction to number fraction.

The size distributions obtained from both methods are shown in Figure 2 on page 12. From the graph we see that both methods yield approximately the same distribution. For this reason and since larger errors were incurred from particle sizes being measured manually from the photographs in the first method, the same distribution obtained from the image analyser was used for all the calculations.

The size distribution used is given numerically in "Appendix K. Feed size distribution" on page 69.

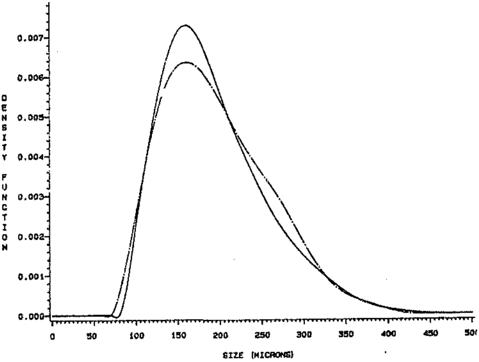

Figure 2. Feed particle size distribution.

Legend: ———————— distribution from photographs

—— · —— distribution from image analyser

3.4 SIZING OF THE TANKS

An approximate value of $d\ell/dt$ was obtained as follows.

A particle was placed in a beaker of ferric sulphate on a magnetic stirrer with a microscope and camera above it. The particle was approximated to an ellipsoid and thus the characteristic length was obtained using:

$$\ell = [\ell_{min}\ell_{max}(\ell_{min} + \ell_{max})\pi/12]^{1/3}$$

where: ℓ_{min} = minimum diameter

ℓ_{max} = maximum diameter

Both ℓ_{min} and ℓ_{max} were measured from the photographs and then scaled to their actual sizes. The results of this photographic technique are shown in Figure 3.

time (min)	size (μm)	dl/dt (μm/min)
0	400	
20	200	10
45	50	6

Figure 3. Particle growth rates.

The average particle size is 200 μm (see Section 3.3) and for the particle mass to be leached by half, the size must be leached by 40 μm. This, according to the values in Figure 3, requires approximately five minutes.

Now by mass balance

$$\tau = V(C_i - C_o)/2CM \qquad ..(1)$$

where: V................tank volume
C_i........inlet ferric conc.
C_o.......outlet ferric conc.
C......fractional conversion
M..............mass flowrate

Consider a typical run where expected values might be:

$V = 1.0$ litre
$C_i = 25.0$ g/litre
$C_o = 10.0$ g/litre
$M = 3.2$ g/min
$C = 0.5$

From equation (1) we get $\tau = 5$ minutes

ie. Using a tank size of one litre yields a residence time of the required magnitude.

3.5 APPARATUS

A schematic diagram of the apparatus is shown in Figure 4.

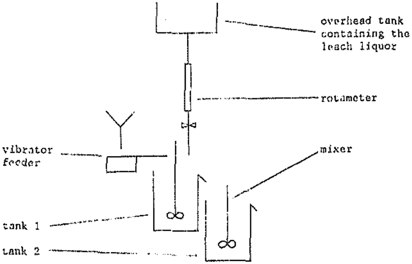

Figure 4. The experimental apparatus

Two vibrator feeders were used in series. The first was used to regulate the solid flowrate and the second placed on its highest setting to ensure an unhindered flow to the tanks. Two flexidrive motors were used to drive the impellers. Rotameters were used to control the liquid flowrate to the tanks.

To ensure that the tanks simulated well-mixed tank a trial run using water and sand was conducted. The concentrations of the sand in water was checked in the tanks and in the outlet steams of the tanks. Results show that these concentrations are consistent with the input concentrations. The results are shown in "*Appendix N. Run with sand and water*" *on page* 73.

3.6 PROCEDURE

The following procedure was used for each run.

The system was run for a period of approximately five residence times to reach steady state.

The vibrator feeder was fed onto a mass balance for twenty minutes before and ten minutes after each run. From this the reliability and average flowrate of the feeder could be calculated. The flowrates proved to be constant. The mass flowrate for the first run is shown in "Appendix F. Mass flowrates of first run" on page 48.

Samples were drawn from each tank and the sample volumes noted. The samples were analysed as follows:

1. the total aqueous iron concentration was obtained by A.A. spectroscopy.

2. the ferric ion concentration was determined by the titration as shown in "Appendix D. Titration for Ferric ion" on page 46.

3. the ferrous ion concentration was determined by the difference of the above concentrations.

4. the H_2SO_4 concentration was determined by the titration as shown in "Appendix E. Titration for Sulphuric acid" on page 47.

4 RESULTS AND ANALYSIS

4.1 COMPARISON OF EXPERIMENTAL TO THEORETICAL RESULTS

The results for the leaching experiments are shown in "Appendix O. Experimental results" on page 74. The results show that the reaction of the acid with the iron was negligible and it was therefore not included in the analysis.

The T vs. C curve (plotted from equation (1) below) for the first tank is unique since the feed particle size distribution is fixed. However the conversions in the first tank were different for each experimental run and this resulted in different inlet particle size distributions and thus different T vs. C curves for the second tank. The problem was therefore how to compare the results for each tank.

It was anticipated that the T vs. C curves for the second tank would not vary greatly from that of the first tank so it was decided to treat the two tanks as two separate single tanks. A program was written to solve the equations (shown below) for each tank and experimental run. The size distribution of the particles leaving the first tank was calculated from the theory and was used as the feed particle size distribution for the second tank. This was necessary since the feed distribution for the first tank could be evaluated using an image analyser (see Section 3.3) but the samples from the first tank could not since they decayed upon drying. The program used the ferric ion concentration as the basis for the mass balance since severe oxidation of the iron samples yielded them unreliable for use as the basis. The program is shown in "Appendix I. Program 3" on page 54 and the results are shown in "Appendix J. Results of program 3" on page 62.

Recall the following equations from Section 2

$$C_1 = [3(\sigma_0/\eta_0^2)T_1 - 6T_1^2 + 6T_1^3/\beta_1](\eta_0^3/\xi_0) \qquad \ldots(1)$$

$$f_1(\ell) = c_1\beta_1\exp(\alpha_1\ell)\int_{\ell}^{\infty}\exp(-\alpha_1\ell')f_0(\ell')d\ell' \qquad \ldots(2)$$

$$1/\beta_1 = 1 - \int_0^{\infty}\exp(-\alpha_1\ell')f_0(\ell')d\ell' \qquad \ldots(3)$$

$$\alpha_1 = 1/\eta_0 T_1 \qquad \ldots(4)$$

where subscript 0 refers to inlet conditions
and subscript 1 refers to outlet conditions

The results show that β is approximately 1 for the experiments and that the ratios of the moments for the second tank do not vary greatly from the ratios in the first tank. The ratios varied the greatest for the fifth experimental run and thus give the T vs. C curve that is the most different from the curve for the first tank. The T vs. C curve for the first tank and the curve for the second tank from the fifth run were plotted on the same graph (see Figure 5 on page 20). The graph shows that the two curves are approximately the same and therefore it was valid to conduct the analysis treating the two tanks as separate single tanks. Furthermore one can say that all the the other T vs. C curves for the second tank lie between these two curves.

Using the fitted kinetics obtained in Section 4.2 one can calculate an experimental value for T as follows.

From the rate equation determined in Section 4.2 and equation (9) from Section 2 one can write

$$T_1 = (\omega\phi/3\rho)(V_1/Q)k_1(Fe^{3+})_1{}^b/\eta_0 \qquad \ldots(5)$$

Using the size distributions calculated from the theory in the program "Appendix I. Program 3" on page 54, and by substituting the following values into equation (5)

$\rho = 7.5 \times 10^6$ g/m^3
$w = 55.85$ g/mol
$\phi = 4.7$

one can calculate T for each tank and experimental run. These experimental T values and their corresponding conversions were plotted against the T vs. C curves shown in Figure 5 on page 20. The plot is shown in Figure 6 on page 21. and from it one can see that there is good agreement with the theory although it must be remembered that fitted kinetics were used.

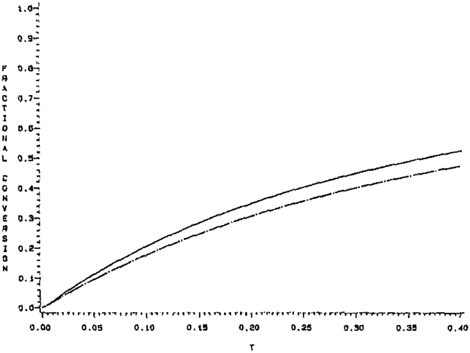

Figure 5. Comparison of T vs. C curves for the first tank and for the second tank and fifth run :

Legend: ———— first tank

—— - —— second tank

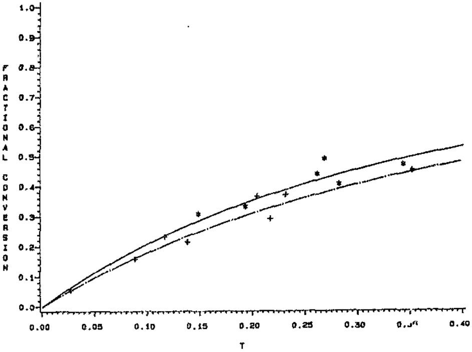

Figure 6. *Comparison of theoretical* T vs. C curves to the experimental T and C values :

Legend: ———— first tank

—— · —— second tank

* experimental first tank points

\+ experimental second tank points

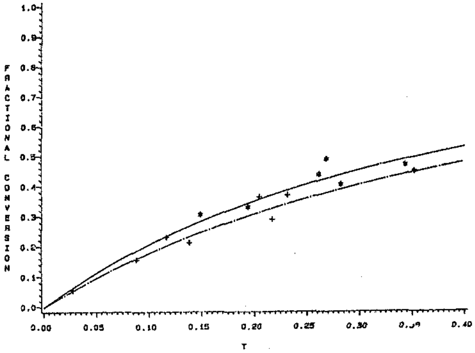

Figure 6. Comparison of theoretical T vs. C curves to the experimental *T and C values :*

Legend: ———————— first tank

——— · ——— second tank

☆ experimental first tank points

\+ experimental second tank points

4.2 THE REACTION KINETICS

The reaction

$$Fe + Fe_2(SO_4)_3 \rightarrow 3FeSO_4$$

is electrochemical in nature and involves the following half reactions

$$Fe^{3+} + e' \rightarrow Fe^{2+}$$

$$Fe \rightarrow Fe^{2+} + 2e'$$

Reaction of the acid with the iron proved to be negligible and was therefore not considered. (see Section 4.1)

Sketches of the current - potential curves for the above half reactions are shown in Figure 7.

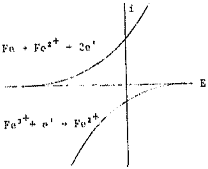

Figure 7. current-potential curves

By modelling these curves the form of the rate equation can be derived. (see "*Appendix B. Form of rate equation*" on page 41)

The form was derived to be the following

$$dm(\ell)/dt = a(\ell)\omega k(Fe^{3+})^b \qquad \ldots(1)$$

where k and b are constants

Values for these constants were determined by considering each tank as a separate single tank with the calculated exit size distribution of the first tank being used as the feed size distribution for the second tank. Values for the constants were evaluated as follows.

Recall equation (3) from Section 2

$$T = (\omega\phi/3\rho)(V/Q)k\psi/\eta$$

Substitution of equation (1) gives

$$T = (\omega\phi/3\rho)(V/Q)k(Fe^{3+})^b/\eta \qquad \ldots(2)$$

where η is the first moment of the feed size distribution

Hence

$$A = \ln(k) + b\ \ln[(Fe^{3+})_i]$$

where: $A = \ln[(3\rho/\phi\omega)(Q/V)\eta T]$
$\rho = 7.5 \times 10^6$ g/m³
$\phi = 4.7$
$\omega = 55.85$ g/mol
Q ....ℓ/min

V .... ℓ

η_e .... m

k .... m/min (for b=1)

The method by which the variables of the system were evaluated for each experimental run are described in section 4.1 "Comparison of Experimental to Theoretical Results" on page 17. An extension to the program was made to evaluate A and $\ln(Fe^{2+})$ from the system's variables and to solve for the reaction constants by linear regression. The results of the program are shown in "Appendix J. Results of program 3" on page 62 and the program is shown in "Appendix I. Program 3" on page 54.

The regression was first used for all the A values and then for the A values from each tank separately. These results are shown in Figure 8.

Tank's used in the regression	Reaction constant	Reaction order
Both tanks	0.00327	1.097
1st tank	0.00513	1.040
2nd tank	0.00345	1.052

Figure 8. table of the reaction kinetics

Figure 8 shows that the reaction is approximately first reaction so it was assumed that the reaction is indeed first order and the regression repeated to find the reaction constant. These results are shown in Figure 9 on page 25.

Tank's used in the regression	Reaction constant m/min	Reaction order
Both tanks	0.00527	1.000
1st tank	0.00630	1.000
2nd tank	0.00439	1.000

Figure 9. table of the reaction kinetics: the reaction constant is assumed to be unity

We see from Figure 9 that each tank has a different reaction constant. This is not entirely unexpected as it likely that the most active reactive *sites are used* during reaction in the first tank resulting in an increased activation energy and thus giving a lower reaction constant in the second tank (Brittan 1976).

Graphs showing the linear regression for Figure 9 are shown in Figure 12 on page 27 , *Figure 10 on page 26 and* Figure 11 on page 26.

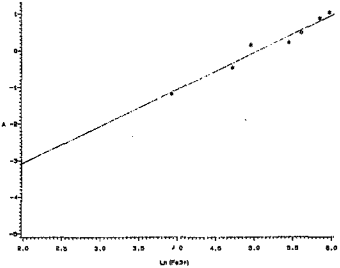

Figure 10. Plot of A vs. ln(Fe+$^{3+}$) : first tank.

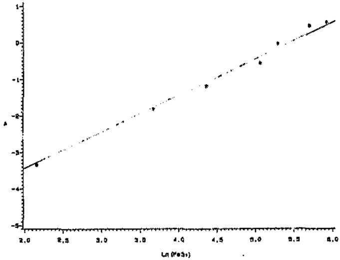

Figure 11. Plot of A vs. ln(Fe+$^{3+}$) : second tank.

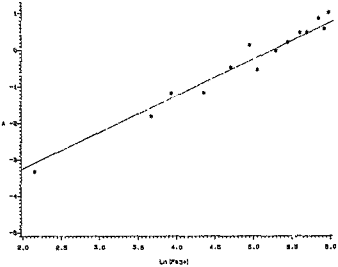

Figure 12. Plot of A vs. $\ln(Fe+^{2+})$: both tanks.

4.3 PROCEDURE FOR THE DESIGN OF TANKS

One can see from the equations in Section 2 that in a two tank system the first tank is described by a unique curve of T_1 vs. C_1 whilst for the second tank the curve of T_2 vs. C_2 is dependant on C_1 and *is therefore* not unique. This means that a one tank system can be easily modelled by a single C vs. T curve whilst an *iterative procedure is required* to solve a two tank system.

Algorithm for a one tank system

The following equations are derived in section 2.

$$C_1 = [3(\sigma_0/\eta_0^2)T_1 - 6T_1^2 + 6T_1^3/\beta_1](\eta_0^3/\xi_0) \qquad \ldots(1)$$

$$T_1 = (\omega\phi/3\rho)(V_1/Q)k_1\psi_1(C_1)/\eta_0 \qquad \ldots(2)$$

$$f_1(\ell) = (\beta_1/\eta_0 T_1)\exp(\ell/\eta_0 T_1)\int_{\ell}^{\infty}\exp(-\ell'/\eta_0 T_1)f_0(\ell')d\ell' \qquad \ldots(3)$$

$$1/\beta_1 = 1 - \int_{0}^{\infty}\exp(-\ell'/\eta_0 T_1)f_0(\ell')d\ell' \qquad \ldots(4)$$

A plot *of C vs. T is made using* equation (1). From this curve the value of T can be obtained for any conversion. Using equation (2) the system variables required to achieve this conversion can be calculated. This assumes that k and $\psi(C)$, which contain the kinetics of the reaction are known. ψ is a function of the reactant concentrations which in turn are functions of the conversion and are linked by a mass balance of the system.

Algorithm for a two tank system

The following equations hold for the i^{th} tank (see Section 2):

$$C_i = [3T_i\sigma_{i-1}/(\eta_{i-1})^2 - 6T_i^2 + 6T_i^3/\beta_i](\eta_{i-1}^3/\xi_{i-1}) \qquad \ldots(1)$$

$$T_i = (\omega\phi/3\rho)(V_i/Q)k_i\psi_i(C_i)/\eta_{i-1} \qquad \ldots(2)$$

$$f_i(\ell) = \beta_i/(\eta_{i-1}T_i)\exp(\ell/\eta_{i-1}T_i)\int_\ell^\infty \exp(-\ell'/\eta_{i-1}T_i)f_{i-1}(\ell')d\ell' \qquad \ldots(3)$$

$$1/\beta_i = 1 - \int_0^\infty \exp(-\ell'/\eta_{i-1}T_i)f_{i-1}(\ell')d\ell' \qquad \ldots(4)$$

From a mass balance over the whole system

$$C = 1 - (1 - C_1)(1 - C_2) \qquad \ldots(5)$$

where: C = fractional conversion over the whole system
C_i = fractional conversion in tank i

From equation (2) with i=1 and i=2 one can derive

$$T_1 - T_2(V_1/V_2)(\eta_1/\eta_0)(k_2/k_1)[\psi(C_1)/\psi(C_2)] = 0 \qquad \ldots(6)$$

Note: if $C_1 = 0$ then $C_2 = C$ and the L.H.S of (6) is negative
if $C_1 = C$ then $C_2 = 0$ and the L.H.S of (6) is positive

i.e. Any root finding method can be used to solve for C_1. Iteration over C_1 has the advantage that C is bounded between 0 and the required conversion whereas iteration over the volume is not bounded. The L.H.S of (6) is evaluated as follows·

1. Guess C_1 and calculate C_2 from (5).

2. Calculate T_1 using an iterative technique for (1) where i=1

3. Calculate $f_1(\ell)$ from (3) and hence calculate the moments.

4. Calculate T_2 using an iterative technique for (1) where i=2

5. Calculate the L.H.S of (6).

Algorithm for N tanks

The following equations hold for the ith tank (see Section 2):

$$C_i = [3T_i\sigma_{i-1}/(\eta_{i-1})^2 - 6T_i^2 + 6T_i^3/\beta_i](\eta_{i-1}^3/\xi_{i-1}) \qquad \ldots(1)$$

$$T_i = (\omega\phi/3\rho)(V_i/Q)k_i\psi_i(C_i)/\eta_{i-1} \qquad \ldots(2)$$

$$f_i(\ell) = \beta_i/(\eta_{i-1}T_i)\exp(\ell/\eta_{i-1}T_i)\int_\ell^\infty \exp(-\ell'/\eta_{i-1}T_i)f_{i-1}(\ell')d\ell' \qquad \ldots(3)$$

$$1/\beta_i = 1 - \int_0^\infty \exp(-\ell'/\eta_{i-1}T_i)f_{i-1}(\ell')d\ell' \qquad \ldots(4)$$

From a mass balance over the whole system

$$C = 1 - (1 - C_1)(1 - C_2)\ldots\ldots(1 - C_N) \qquad \ldots(5)$$

where: C = fractional conversion over the whole system
C_i = fractional conversion in tank i

One can solve the system for the required variable by iterating around that variable. For example if one wanted the tank volumes required to give a certain conversion then one would iterate around the volume, but the ratios of the tank volumes must be stipulated beforehand. We cannot solve the system by iterating around the intermediate conversions as in the case for two tanks since there is no relationship between these conversions.

This is disadvantageous since it increases the amount of iterations, but it must be realised that few industrial systems use more than three tanks. For each iteration the system is solved as follows.

1. Evaluate equations (1) through (4) for each of the tanks noting that ψ is a function of the reaction kinetics and the mass balance.

2. Evaluate the fractional conversion from equation (5).

3. Select a new value for the variable.

5 CONCLUSIONS AND RECOMMENDATIONS

The particle number balance approach to the modelling of leaching in well-mixed tanks, as advocated in this work, is a novel and valid approach. This is indicated by the leaching experiments which were successfully modelled. However, at present the model is limited to particles which exhibit the shrinking particle behaviour upon leaching. This mechanism is characterised by two main features. The first is that the particles are non-porous so that reaction only occurs at the surface and the second is that the leached material is released from the particle without affecting the rate. Many reactions have these characteristics but it should not be difficult to extend this model to other mechanism.

The model was developed from a particle number balance over the i^{th} tank in a series of n well-mixed tanks. The balance gave a first order differential equation which was solved to give the size distribution and the ratio of the number of particles entering to those exiting the i^{th} tank.

Two major conclusions can be drawn from the theory and the experiments. The first is that a simple leach reaction with fitted kinetics in a single tank can be modelled. The leach efficiency can, for a given feed particle size distribution, be modelled by a unique dimensionless graph. This graph is a plot of the leach efficiency versus a dimensionless number (T), which incorporates the reaction constants and system variables. From the graph, T can be found for a given conversion and thus the volume or flowrate to give this conversion can be calculated. The algorithm to solve a one tank system is shown in Section 4.3.

The second conclusion is that the graphical technique is not easily extended to incorporate multiple tanks. This is because a unique curve of conversion versus T cannot be plotted for consecutive tanks since the curve is a function of the conversion in the previous tanks. A system

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The second conclusion is that the graphical technique is not easily extended to incorporate multiple tanks. This is because a unique curve of conversion versus T cannot be plotted for consecutive tanks since the curve is a function of the conversion in the previous tanks. A system incorporating multiple tanks must therefore be evaluated using an iterative procedure. To obtain the solution for the two tank system one can iterate using the conversion over the first tank. This is advantageous since the conversion is bounded between 0 and 1. The N tank system however, requires iteration over the desired variable eg. volume, which is disadvantageous since this variables is unbounded. The calculations get progressively more extensive as more tanks are considered but most processes only use a maximum of three tanks. Examples of this are in the leaching of zinc and manganese and the pressure leaching of nickel, copper and cobalt mattes. The algorithms for the two and N tank systems are shown in Section 4.3.

Further work be conducted in extending the model to incorporate non-uniform dissolution mechanisms (ie. where $d\ell/dt$ is a function of ℓ). This would remove some of the limitations on the model and make it more general.

An attempt could be made to optimise the conversion with respect to the number and volume of the tanks required. This would, however, be very complicated since a numerical search technique would have to be devised, based on an objective function which would include costs of equipment and operation.

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APPENDIX A. DETAILED THEORY

If we assume the ideal shrinking particle model then

$$\begin{aligned} dm(\ell)/dt &= -\, k_i \psi_i a(\ell) \omega \\ &= -\, k_i \psi_i \phi \ell^2 \omega \\ &= 3\rho \ell^2 u_i \omega \end{aligned}$$

Therefore

$$u_i = -k_i \psi_i \phi \omega / 3\rho \qquad \ldots(1)$$

where:

$a(\ell)$ = exposed surface area of particle of size ℓ
k_i = rate constant in tank i
ℓ = characteristic particle size
= $[m(\ell)/\rho]^{1/3}$
$m(\ell)$ = mass of single particle of size ℓ
t = residence time of particle in contact with solution
u_i = particle growth rate in tank i
= $d\ell/dt$
ϕ = shape factor
= $a(\ell)/\ell^2$
ψ = quantity dependant on chemical and physical properties of the solution environment in tank i
ρ = density of particle
ω = molecular weight of dissolving species

i.e. the ideal shrinking particle model assumes u_i is independent of ℓ.

Consider a particle balance over the i^{th} tank.

And assuming :

1. steady state.
2. well mixed tanks
3. Q is constant.
4. ideal shrinking particle model.

CONVECTION + BIRTH = DEATH

$$N_i V_i u_i \, df_i(\ell)/d\ell + N_{i-1} Q f_{i-1}(\ell) = N_i Q f_i(\ell) \qquad \ldots(2)$$

where:

$f_i(\ell)d\ell$ = fraction of particles in tank i with sizes between ℓ and $\ell+d\ell$

N_i = no. of particles in tank i per unit volume of solution

Q = volumetric flowrate of solution

V_i = volume of tank i

Define:

$$\alpha_i = -Q/u_i V_i \qquad \ldots(3)$$

and

$$\beta_i = N_{i-1}/N_i \qquad \ldots(4)$$

Substitution of (3) and (4) in (2) gives

$$df_i(\ell)/d\ell - \alpha_i f_i(\ell) = - \beta_i \alpha_i f_{i-1}(\ell) \qquad \ldots(5)$$

integrating (5)

$$f_i(\ell) = \exp(\alpha_i \ell) f_i(0) - \exp(\alpha \ell) \int_0^{\ell} \beta_i \alpha_i \exp(-\alpha_i \ell') \, f_{i-1}(\ell') d\ell' \qquad \ldots(6)$$

Equation (6) must satisfy the following conditions:

1. as $\ell \to \infty$ so $f_i(\ell) \to 0$

2. $\int_0^{\infty} f_i(\ell) d\ell = 1$

Now α_i is +ve so $\exp(\alpha_i \ell) \to 0$ as $\ell \to \infty$

$$f_i(0) = \int_0^{\infty} \beta_i \alpha_i \exp(-\alpha_i \ell') f_{i-1}(\ell') d\ell' \qquad \ldots(7)$$

substituting in (6)

$$f_i(\ell) = \beta_i \alpha_i \exp(\alpha_i \ell) \int_0^{\ell} \exp(-\alpha_i \ell') f_{i-1}(\ell') d\ell' \qquad \ldots(8)$$

Using condition (2) for equation (8)

$$\int_0^\infty f_i(\ell) = -\beta_i[1 - \int_0^\infty \exp(-\alpha_i \ell')f_{i-1}(\ell')d\ell']$$

therefore

$$\beta_i = 1/[1 - \int_0^\infty \exp(-\alpha_i \ell')f_{i-1}(\ell')d\ell'] \qquad \ldots(9)$$

Using (9) we can derive the following

$$\eta_i = \int_0^\infty \ell\, f_i(\ell)d\ell = \beta_i \eta_{i-1} - 1/\alpha_i \qquad \ldots(10)$$

$$\sigma_i = \int_0^\infty \ell^2 f_i(\ell)d\ell = \beta_i \sigma_{i-1} - 2\eta_i/\alpha_i \qquad \ldots(11)$$

$$\xi_i = \int_0^\infty \ell^3 f_i(\ell)d\ell = \beta_i \xi_{i-1} - 3\sigma_i/\alpha_i \qquad \ldots(12)$$

therefore ξ_i can be obtained from η_0 , σ_0 and ξ_0

If $$M_i = \rho Q N_i \int_0^\infty \ell^3 f_i(\ell)d\ell \qquad \ldots(13)$$

Then the fractional conversion C is given by

$$C = 1 - M_n/M_0$$

$$= 1 - \prod_{i=1}^{n} M_i/M_{i-1}$$

$$= 1 - \prod_{i=1}^{n} (1/\beta_i)(\xi_i/\xi_{i-1})$$

$$= 1 - (\xi_n/\xi_0) \prod_{i=1}^{n} 1/\beta_i \qquad \ldots(14)$$

The equations are used as follows for conversion in one tank

Define $\tau_i = V_i/Q$

$$t_i^* = -\eta_0/u_i \qquad \ldots(15)$$

$$T_i = \tau_i/t_i^* \qquad \ldots(16)$$

Then from (3)

$$\alpha_i = 1/\eta_0 T_i \qquad \ldots(17)$$

equations (10) ,(11) and (12) give

$$\eta_i = \beta_i\eta_0 - 1/\alpha_i \qquad \ldots(18)$$

$$\sigma_i = \beta_i\sigma_0 - 2\beta_i\eta_0/\alpha_i + 2/(\alpha_i)^2 \qquad \ldots(19)$$

$$\xi_i = \beta_i\xi_0 - 3\beta_i\sigma_0/\alpha_i + 6\beta_i\eta_0/(\alpha_i)^2 - 6/(\alpha_i)^3 \qquad \ldots(20)$$

Substituting (17) and (20) into (14) gives

$$C_i = [3(\sigma_0/\eta_0^2)T_i - 6T_i^2 + 6T_i^3/\beta_i](\eta_0^3/\xi_0) \qquad \ldots(21)$$

From equations (1) , (15) and (16) we have

$$T_i = (\omega\phi/3\rho)(V_i/Q)k_i\psi_i/\eta_0 \qquad \ldots(22)$$

recall equation (9)

$$1/\beta_i = 1 - \int_0^\infty \exp(-\alpha_i \ell')f_{i-1}(\ell')d\ell'$$

The size distribution is obtained from (8) and is given by

$$f_1(\ell) = \alpha_1\beta_1\exp(\alpha_1\ell)\int_\ell^\infty \exp(-\alpha_1\ell')f_0(\ell')d\ell' \qquad \ldots(23)$$

where $\alpha_1 = 1/\eta_s T$

APPENDIX B. FORM OF RATE EQUATION

The Butler-Volmer (Pickett, 1979) equation is as follows

$$i = i_0[\exp((1-\beta)zF/RT\eta - \exp(-\beta zF/RT\eta)] \qquad \ldots(1)$$

where: F = Faraday's constant
i = current density
i_0 = exchange current density
R = gas constant
T = temperature
z = no. of electrons transferred
β = symmetry factor
η = over potential

The reaction

$$Fe \rightarrow Fe^{3+} + 2e'$$

is in the anodic region where η is large and positive so the Butler-Volmer equation reduces to

$$i = i_0\exp((1-\beta)zF/RT\eta \qquad \ldots(2)$$

The Nernst equation (Bockriss, 1970) gives

$$\eta = E - [E^0 + (RT/2F)\ln(Fe^{2+})] \qquad \ldots(3)$$

where: E = potential

E^0 = standard equilibrium potential

Now the following has been previously verified

$$i_0 = i_0^{*}(Fe^{2+})^{\frac{1}{2}} \qquad \ldots(4)$$

where: i_0^{*} = exchange current density independent of concentration

From equation (2) , (3) and (4) we get

$$i_1 = h_1 \exp(-a_1 E) \qquad \ldots(5)$$

where: $a_1 = -F/RT$

$h_1 = i_0^{*} \exp(a_1 E)$

similarly for the reaction

$$Fe^{3+} + e^{-} \rightarrow Fe^{2+}$$

we have

$$i = -i_0 \exp(-\beta z F/RT\eta) \qquad \ldots(6)$$

$$\eta = E - [E^{\circ} + (RT/F)\ln(Fe^{3+}/Fe^{2+})] \qquad \ldots(7)$$

$$i_0 = i_0^{*}(Fe^{3+})^{\frac{1}{2}}(Fe^{2+})^{\frac{1}{2}} \qquad \ldots(8)$$

$$i_2 = -h_2(Fe^{3+})\exp(-a_2 E)$$

where: $a_2 = -F/2RT$ ...(10)

$h_2 = i_0^{*} \exp(a_2 E)$

However at large overpotentials the reaction is known to be mass transfer controlled and thus takes the form

$$i_2 = -k(Fe^{3+}) \qquad \ldots(11)$$

Since there is no nett transfer of current we have

$$i_1 + i_2 = 0 \qquad \ldots(12)$$

Using Faraday's Law and by considering a particle of size ℓ we can say

$$dm(\ell)/dt = a(\ell)\omega i_1/2F \qquad \ldots(13)$$

where: $a(\ell)$ = exposed area of particle size ℓ
F = Faraday's constant
ω = molecular weight of dissolving species.

So using equations (5) , (9) , (12) and (13) we have:

For the reaction not mass transfer controlled

$$dm(\ell)/dt = a(\ell)\omega k(Fe^{3+})^{2/3}$$

For the reaction mass transfer controlled

$$dm(\ell)/dt = a(\ell)\omega k(Fe^{3+})$$

i.e. the rate equation has the following form

$$dm(\ell)/dt = a(\ell)\omega k(Fe^{3+})^{b}$$

where k = some constant
b = some constant

APPENDIX C. OXIDATION OF FERROUS SULPHATE

The following molecular masses were used:

$FeSO_4.7H_2O$ = 278.05 g/mol
$Fe_2(SO_4)_3$ = 111.75
Fe = 55.85
H_2SO_4 = 98.07
H_2O_2 = 34.01

the specific gravities of 98% H_2SO_4 = 1.84
30% H_2O_2 = 1.11

Ferrous sulphate is oxidised as :

$$2FeSO_4 + H_2 + H_2O_2 = Fe_2(SO_4)$$

Now 1/111.7 mole of $Fe_2(SO_4)_3$ contains 1 g of Fe^{3+}

To ensure that all the peroxide reacted 3/4 the stoichiometric amount was added. By experiment it was found that for concentrations of iron higher than 40 g/l there was extensive complexing of the iron which was indicated by the dark red colour of the solution.

The storage vessel for the ferric sulphate had a capacity of 35 litres and it was found that the following reagents were needed.

35 litres	H_2O
7 kg	$FeSO_4.7H_2O$
680 ml	98% H_2SO_4
1 litre	30% H_2O_2

The solution was analysed as follows:

1. the total aqueous iron concentration was obtained by A.A. spectroscopy.
2. the ferric ion concentration was determined by the titration as shown in "Appendix D. Titration for Ferric ion" on page 46.
3. the ferrous ion concentration was determined by the difference of the above two concentrations.
4. the H_2SO_4 concentration was determined by the titration as shown in "Appendix D. Titration for Ferric ion" on page 46.

APPENDIX D. TITRATION FOR FERRIC ION

Prepared solutions

A standard 0,179 N sodium thiosulphate solution was prepared by dissolving 44,424 g $Na_2S_2O_3.5H_2O$ in recently boiled distilled water and making up to 1,0 litres. Approximately 3 drops of chloroform and 0,1 g/litre Na_2CO_3 was added.
Then 1,0 ml 0,179 N $Na_2S_2O_3$ = 0.01 g Fe^{3+}

Method

1. Add an excess of KI crystals to an aliquote of the leach sample.

2. Titrate with the standard sodium thiosulphate solution until the range brown colour disappears.

3. Near the end point the solution becomes pale yellow, and at this point a crystal of KI may be added to determine whether sufficient KI is present.

4. At the end point the solution is colourless.

5. Calculate the concentration of Fe^{3+} in the original leach solution.

Reactions

$Fe_2(SO_4)_3 + KI \rightarrow 2FeSO_4 + I^0$

$I^0 + Na_2S_2O_3 \rightarrow Na_2S_4O_6 + NaI$
(colourless)

APPENDIX E. TITRATION FOR SULPHURIC ACID

Prepared solutions

1. Mixed indicator : 0,2 g dimethyl yellow and 0,2 g methyl blue dissolved in 200,0 ml methanol.

2. *0,204 N Sodium carbonate prepared by dissolving 10,8110 g oven dried* (at 533,0 K -543,0 K) Na_2CO_3 with distilled water up to 1,0 litres.

Then 1 ml 0,203 N Na_2CO_3 = 0,01 g H_2SO_4

Method

1. Any ferric ion present in an aliquote of the leach solution must first be reduced to ferrous ion (i.e. add KI crystals and titrate with $Na_2S_2O_3$ as in "Appendix D. Titration for Ferric ion" on page 46)

2. Add 3 drops mixed indicator to the aliquote and titrate with the *standard Na_2CO_3 solution until a green colour indicates that the end* point is reached.

Reaction

$Na_2CO_3 + H_2SO_4 \rightarrow Na_2SO_4 + CO_2 + H_2O$

APPENDIX F. MASS FLOWRATES OF FIRST RUN

Time (min)	Mass flowrate (g/min)
before run	
1	3,24
2	3,26
3	3,23
4	3,20
5	3,18
6	3,20
7	3,10
8	3,05
9	3,05
10	2,98
11	3,05
12	3,11
13	3,15
14	3,20
15	3,20
16	3,25
17	3,20
18	3,18
19	3,18
20	3,16
after run	
1	3,26
2	3,30
3	3,28
4	3,24
5	3,18
6	3,20
7	3,16
8	3,14
9	3,16
10	3,16

APPENDIX G. PROGRAM 1

The particles were approximated to ellipsoids and their maximum and minimum dimensions measured. The particles were photographed on millimetre graph paper and thus the scale could be determined.

The algorithm

Initialisation

Input of data

By assuming the particles were ellipsoids the volume and area were calculated. Hence:

characteristic length = $(\text{volume})^{1/3}$

shape factor = $\text{area}/(\text{characteristic length})^{1/2}$

This was done for each particle

The particles were tested to see which size interval they fell in and were noted as such.

A cubic spline was fitted to the intervals and interpolated to yield the size distribution .

The program

```
C      INITIALISATION
       DIMENSION D(500),PH(200),DF(500),DL(500)
       DIMENSION F(100),XL(100),YF(100),COEF(100,3)
       DIMENSION WK(2000),YYF(100)
       REAL L(500)
C
       PI=3.1416
C      SET PARAMETERS FOR IMSL ROUTINES
       IJOB=1
       IC=100
C      NP IS THE NO. OF PARTICLES
       NP=180
C
C      INPUT OF DATA
       NN=2*NP
       READ(9,*)(D(I),I=1,NN)
C
       DO 10 J=2,NN,2
         I=J/2
C        A AND B ARE MAX AND MIN DIMENSIONS OF PARTICLE
         SCALE=20.
         A=D(J-1)*SCALE
         B=D(J)*SCALE
         C=A/B
C        CALC. OF CHARACTERISTIC LENGTH
C        (ASSUMES PARTICLES ARE ELLIPSOIDS)
```

```
        DL(I)=(PI*A*B*(A+B)/12.)**(1/3.)
        IF(A.NE.B)THEN
          C2=(C**2.-1.)**0.5
          AREA=(PI/2)*A*B/C2*(ALOG(C+C2)+C*ATAN(C2))
        ELSE
          AREA=PI*A**2.
        END IF
C       CALC. OF SHAPE FACTOR
        PH(I)=AREA/DL(I)**2
10    CONTINUE
C
      RMIN=100.
      NI=10
      H=50.
C     EVALUATION OF DISCRETE F(L)
      DO 50 I=1,NI
        LLIMIT=(I-1)*H
        ULIMIT=(I)*H
        DO 60 J=1,NP
          DD=DL(J)
          IF(DD.GT.RMIN.AND.DD.GE.LLIMIT.AND.DD.LT.ULIMIT)THEN
              DF(I)=DF(I)+1
              PHI=PHI+PH(J)
            END IF
60      CONTINUE
        DFTOT=DFTOT+DF(I)
50    CONTINUE
C
      DO 70 I=1,NI
        DF(I)=DF(I)/DFTOT
70    CONTINUE
      PHI=PHI/DFTOT
C
C     CALC. DENSITY DISTRIBUTION POINTS FOR ICSCUU WHICH CALCULATES
C     THE CUBIC SPLINE COEFFICIENTS
      NX=NI+2
      DO 100 K=2,NI+1
        XL(K)=(K-1.5)*H
        YYF(K)=DF(K-1)/H
100   CONTINUE
      XL(NX)=(NX-2)*H
C
C     CALL SUB TO EVALUATE SPLINE COEFFICIENTS
      CALL ICSSCV(XL,YYF,NX,YF,COEF,IC,IJOB,WK,IER)
C
C     CALC OF NEW NUMBER OF POINTS AND THEIR VALUES FOR L
      HNEW=10.
      N=XL(NX)/HNEW+1
      H=HNEW
      DO 110 K=1,N
110   L(K)=(K-1)*H
C
C     EVALUATION OF F(L) FOR THE VALUES OF L
      CALL ICSEVU(XL,YF,NX,COEF,IC,L,F,N,IER)
```

```
      F(1)=0.0
      DO 115 I=2,N
115   F(I)=AMAX1(F(I),0.0)
      F(N)=0.0
C
C     NORMALISE F(L)
      CALL ICSCCU(L,F,N,COEF,IC,IER)
      CALL DCSQDU(L,F,N,COEF,IC,L(1),L(N),SUM,IER)
      DO 120 K=1,N
120   F(K)=F(K)/SUM
C
C     PRINTING OF THE SHAPE FACTOR
      WRITE(10,150)PHI
150   FORMAT(//,'THE SHAPE FACTOR =',F4.1,//)
C
C     PRINTING OF SIZE DISTRIBUTION
      WRITE(10,*)N,H
      DO 130 K=1,N
        WRITE(10,140)L(K),F(K)
140     FORMAT(5X,F5.1,10X,F9.7)
130   CONTINUE
      STOP
      END
```

APPENDIX H. PROGRAM 2

This program evaluates the size distribution from results of the image analyser. The image analyser's results are in the form of graphs of particle size vs. the mass fraction passing that size.

The algorithm

Initialisation

Input of the data from the graphs which is then averaged.

A cubic spline is fitted the data and the mass distribution interpolated from it.

The mass distribution is converted to a number distribution and normalised.

The program

```
C     INITIALISATION
      DIMENSION F(100),X(100),Y(100),COEF(100,3),P(5,100),FP(100)
      DIMENSION VAL(100),WK(2000),YY(100)
      REAL L(500)
C
C     SET PARAMETERS FOR IMSL ROUTINES
      IJOB=1
      IC=100
C
C     INPUT OF DATA
      READ(15,*)NP,H,NE
      DO 10 I=1,NP
        READ(15,*)(P(J,I),J=1,NE)
C
C       AVERAGING THE READINGS
        FP(I)=0
        DO 20 J=1,NE
          FP(I)=FP(I)+P(J,I)
20      CONTINUE
        FP(I)=FP(I)/NE
C
10    CONTINUE
C
C     CALC. DATA POINTS FOR ICSSCV
      NX=NP+1
      X(1)=0
      YY(1)=0
      DO 40 K=2,NP
        X(K)=(K-1.5)*H
        YY(K)=(FP(K)-FP(K-1))/H
40    CONTINUE
      XMAX=(NX-2)*H
```

```
      X(NX)=XMAX
      YY(NX)=0
C
C     CALL SUB TO EVALUATE SPLINE COEFFICIENTS
      CALL ICSSCV(X,YY,NX,Y,COEF,IC,IJOB,WK,IER)
C
C     SET REQUIRED SIZE DISCRETISATIONS
      HNEW=10.
C     CALC OF THE NEW NO. OF POINTS AND VALUES FOR L
      N=XMAX/HNEW+1
      H=HNEW
      DO 50 K=1,N
50    L(K)=(K-1)*H
C
C     EVALUATION OF F(L) FOR THE VALUES OF L
      CALL ICSEVU(X,Y,NX,COEF,IC,L,F,N,IER)
C
C     CONVERSION TO NUMBER FRACTION
      F(1)=0
      DO 60 K=2,N
        F(K)=AMAX1(0.0,F(K)/L(K)**3)
        IF(L(K).LE.50.0)F(K)=0.0
60    CONTINUE
      F(N)=0
C
C     NORMALISE F(L)
      CALL ICSCCU(L,F,N,COEF,IC,IER)
      CALL DCSQDU(L,F,N,COEF,IC,L(1),L(N),SUM,IER)
      DO 90 K=1,N
90    F(K)=F(K)/SUM
C
C     OUTPUT OF DISTRIBUTION
      WRITE(10,*)N,H
      DO 110 K=1,N
        WRITE(10,120)L(K),F(K)
120     FORMAT(5X,F5.1,10X,F9.7)
110   CONTINUE
C
      STOP
      END
```

APPENDIX I. PROGRAM 3

The algorithm

```
Initialisation

Input of size distribution

Calc. of first three moments and their ratios

DO LOOP for each experimental run

    Reading in of experimental results

    DO LOOP for each tank

        Calc. of conversion and expected ferrous concentration and mass
        flowrates for the given ferric ion concentrations.

        Calc. of T from the equation
        $C_1 = [3\eta_0\sigma_0 T - 6(\eta_0)^3 T^2 + 6(\eta_0)^3 T^3/\beta_1]/\xi_0$
        For certain values of T , C had to be expanded using a Taylor
        series to eliminate rounding errors.

        Calc. of $f_1(\ell)$ from T.
        $\alpha$ and $\beta$ were calculated during the intermediate steps.

        Calc. of the first three moments and their ratios.

        Calc. of A from:
        $A=\ln[(3\rho/\phi\omega)(Q/V)\eta(\tau/t^*)]$
        and calc. of $\ln(Fe^{3+})$

    END OF LOOP for each tank

END OF LOOP for the experimental runs

Calc. of kinetics. This is first done using linear regression on both
tanks and then separately on each tank.
```

The program

```
C      INITIALIS1NG OF PROGRAM
       COMMON /BLOC1/L,F,N,NT,H,M,T,CONV,C,MASS,Q
       DIMENSION C(5),C2(5),CC(5),A(5),F(5,500),VAL(500)
       DIMENSION T(5),CONV(5),COEF(500,3),RMASS(5),RC2(5)
       DIMENSION YPLOT(5,10),XPLOT(5,10),RATIO1(5),RATIO2(5)
       DIMENSION ALPHA(5),BETA(5)
       REAL L(500),MASS(5),M(5,500)
       EXTERNAL FOXC
C
C      DEFINE THE PARAMETERS FOR THE IMSL ROUTINES
```

```
      EPS=0.
      NSIG=6.
      IC=500
C
C     READING IN OF THE FEED SIZE DISTRIBUTION
      READ(10,*)N,H
      DO 10 I=1,N
10    READ(10,*)L(I),F(1,I)
C
C     CALCULATION OF THE FIRST THREE MOMENTS OF THE FEED AND
C     THEIR RATIOS
      NT=1
      DO 40 I=1,3
40    CALL MOM(L,F,N,H,NT,I,M(NT,I))
      CALL RATIO(NT,M,RATIO1,RATIO2)
C
C     DO LOOP TO EXECUTE THE PROGRAM FOR EACH EXPT. RUN
      NRUN=7
      DO 100 K=1,NRUN
C
C       READING IN OF THE SYSTEMS VARIABLES
        READ(11,*)IRUN,MASS(1),C(1),A(1),C2(1),Q,
     #                 MASS(2),C(2),A(2),C2(2),
     #                 MASS(3),C(3),A(3),C2(3),
C
C       MEASURED EXPERIMENTAL VALUES ARE ASSIGNED TO DUMMY VARIABLES
        DO 50 NN=1,3
          RMASS(NN)=MASS(NN)
          RC2(NN)=C2(NN)
          CC(NN)=C(NN)+C2(NN)
50      CONTINUE
C
C       DO LOOP FOR THE TANKS. SUBSCRIPT 1 REFERS TO THE FEED,
C       2 TO THE 1ST TANK AND 3 TO THE 2ND TANK.
        DO 70 NT=2,3
C
C         CALC. OF TAU/T* AND CONVERSION FOR THE GIVEN FE3+ CONC.
          CONV(NT)=Q/2.*(C(NT-1)-C(NT))/MASS(NT-1)*100.
          MAXFN=100
          RLIM1=0.0
          RLIM2=1.0
          CALL ZBRENT(FOXC,EPS,NSIG,RLIM1,RLIM2,MAXFN,IER)
          T(NT)=RLIM2
C
C         CALC. OF VARIABLES FOR TANK NT
          C2(NT)=C2(NT-1)+(C(NT-1)-C(NT))*1.5
          MASS(NT)=(1.-CONV(NT)/100.)*MASS(NT-1)
C
C         CALC. OF F(L) FOR TANK NT
          CALL FOL(L,F,N,NT,H,M,T(NT),ALPHA,BETA,IRUN)
C
C         CALCULATE THE FIRST THREE MOMENTS IN TANK NT AND
C         RATIOS
```

```
          DO 80 I=1,3
80        CALL MOM(L,F,N,H,NT,I,M(NT,I))
          CALL RATIO(NT,M,RATIO1,RATIO2)
C
C         CALC. OF POINTS FOR PLOT TO OBTAIN KINETICS
          PHI=4.7
          V=1.0
          XPLOT(NT,K)=ALOG(C(NT)/55.85E-3)
          YPLOT(NT,K)=ALOG(Q*M(NT-1,1)*T(NT)/(2.4822*V*PHI))
C
70      CONTINUE
C       END OF LOOP FOR THE TANKS
C
C       PRINTOUT ROUTINE
        CALL PRINT(Q,C,C2,RC2,CC,A,MASS,RMASS,T,CONV,M,
     #  IRUN,XPLOT,YPLOT,RATIO1,RATIO2,ALPHA,BETA,NRUN)
C
100   CONTINUE
C     END OF LOOP FOR THE EXPT. RUNS
C
C     CALC. OF REACTION KINETICS
      CALL REACT(NRUN,XPLOT,YPLOT)
C
      STOP
      END
C
C     END OF MAIN PROGRAM
C
C     FUNCTION TO EVALUATE THE DIFFERENCE IN THE CONVERSION
C     CALCULATED FROM THE GIVEN T AND THE CONVERSION
C     CALCULATED FROM THE SYSTEM'S CONCENTRATIONS.
C     (THIS IS FOR THE SUBROUTINE ZBRENT)
C     ---------------------------------------------------
      FUNCTION FOXC(A)
      COMMON /BLOC1/L,F,N,NT,H,M,T,CONV,C,MASS,Q
      DIMENSION C(5),F(5,500),T(5),CONV(5)
      REAL L(500),MASS(5),M(5,500)
C
      T(NT)=A
C     CALL CONVER TO CALC. CONV FOR THE GIVEN T
      CALL CONVER(L,F,N,NT,H,M,T(NT),CCONV,NOTE)
      FOXC=CONV(NT)-CCONV
C
      RETURN
      END
C
C
C     SUBROUTINE TO CALC. ORDER AND RXN CONSTANT BY LINEAR REGRESSION
C     ---------------------------------------------------------------
      SUBROUTINE REACT(NRUN,XPLOT,YPLOT)
      DIMENSION XPLOT(5,10),YPLOT(5,10),X(3),XX(3),XY(3),Y(3)
C
      DO 5 I=2,3
        DO 10 K=1,NRUN
```

```
          X(I)=X(I)+XPLOT(I,K)
          XX(I)=XX(I)+XPLOT(I,K)**2
          Y(I)=Y(I)+YPLOT(I,K)
          XY(I)=XY(I)+XPLOT(I,K)*YPLOT(I,K)
10      CONTINUE
5     CONTINUE
      X(1)=X(2)+X(3)
      XX(1)=XX(2)+XX(3)
      Y(1)=Y(2)+Y(3)
      XY(1)=XY(2)+XY(3)
      NN=NRUN*2
C
C     REACTION KINETICS USING BOTH TANKS
      ORDER=1.0
      RCONST=EXP((Y(1)-X(1))/NN)
      WRITE(14,*)ORDER,RCONST
C
C     REACTION KINETICS OVER EACH TANK
      DO 50 I=2,3
        ORDER=1.0
        RCONST=EXP((Y(I)-X(I))/NRUN)
        WRITE(14,*)ORDER,RCONST
50    CONTINUE
C
      RETURN
      END
C
C
C     SUBROUTINE FOR CONVERSION
C     -------------------------
      SUBROUTINE CONVER(L,F,N,NT,H,M,T,CONV,NOTE)
      DIMENSION F(5,500),VAL(500),COEF(500,3)
      REAL L(500),M(5,500)
      IC=500
C
      IF(T.LE.0.001)THEN
        CONV=0
      ELSE
C       CONVERSION USING TAYLOR SERIES
        SUM=0
        COEF1=(T*M(NT-1,1))**3.*6./M(NT-1,3)
        COEF2=-1./(M(NT-1,1)*T)
        DO 10 I=4,21
C         CALC. OF I TH MOMENT
          CALL MOM(L,F,N,H,NT-1,I,M(NT-1,I))
          FACT=1
          DO 30 K=1,I
30        FACT=FACT*K
          SUM1=COEF2**I/FACT*M(NT-1,I)
          SUM=SUM+SUM1
          IF(ABS(SUM1*COEF1*100).LE.0.1E-6)GOTO 40
10      CONTINUE
40      LINE=40
        CONV=(1.-COEF1*SUM)*100.
```

```
        NOTE=I
C
C       CONV BY ANALYTICAL METHOD
        IF(NOTE.GT.20)THEN
          COEF1=3.*M(NT-1,1)*M(NT-1,2)/M(NT-1,3)
          COEF2=6.*M(NT-1,1)**3/M(NT-1,3)
          COEF3=-1./(M(NT-1,1)*T)
          DO 50 J=1,N
           COEF4=AMAX1(-30.0,L(J)*COEF3)
50        VAL(J)=EXP(COEF4)*F(NT-1,J)
          CALL SIMP(N,H,VAL,RINT)
          CONV=(COEF1*T-COEF2*T**2+COEF2*(1.-RINT)*T**3)*100.
        END IF
      END IF
C
      RETURN
      END
C
C
C     SUBROUTINE TO EVALUATE F(L)
C     ---------------------------
      SUBROUTINE FOL(L,F,N,NT,H,M,T,ALPHA,BETA,IRUN)
      DIMENSION F(5,500),VAL(500),COEF(500,3),ALPHA(5),BETA(5)
      REAL L(500),M(5,500)
      IC=500
C
      ALPHA(NT)=1./(M(NT-1,1)*T)
C
C     CALC. OF BETA
      DO 10 J=1,N
        COEF1=AMAX1(-30.0,-ALPHA(NT)*L(J))
        VAL(J)=EXP(COEF1)*F(NT-1,J)
10    CONTINUE
      CALL ICSCCU(L,VAL,N,COEF,IC,IER)
      CALL DCSQDU(L,VAL,N,COEF,IC,L(1),L(N),RINT,IER)
      BETA(NT)=1.0/(1.0-RINT)
C
      DO 20 K=1,N
        DO 30 J=K,N
          COEF3=AMAX1(-30.0,ALPHA(NT)*(L(K)-L(J)))
          VAL(J)=EXP(COEF3)*F(NT-1,J)
30      CONTINUE
C
        CALL ICSCCU(L,VAL,N,COEF,IC,IER)
        CALL DCSQDU(L,VAL,N,COEF,IC,L(K),L(N),RINT,IER)
        F(NT,K)=BETA(NT)*ALPHA(NT)*RINT
        F(NT,K)=AMAX1(0.0,F(NT,K))
20    CONTINUE
C
C     NORMALISING F(L)
      DO 40 I=1,N
40    VAL(I)=F(NT,I)
      CALL SIMP(N,H,VAL,SUM)
```

```
      DO 50 J=1,N
50    F(NT,J)=F(NT,J)/SUM
C
      RETURN
      END
C
C
C     SUBROUTINE TO CALCULATE MOMENTS
C     -------------------------------
      SUBROUTINE MOM(L,F,N,H,NT,I,RMOM)
      DIMENSION F(5,500),VAL(500)
      REAL L(500)
C
      DO 10 J=1,N
10    VAL(J)=F(NT,J)*L(J)**(I*1.0)
      CALL SIMP(N,H,VAL,RMOM)
C
C     CALL ICSCCU(L,VAL,N,COEF,IC,IER)
C     CALL DCSQDU(L,VAL,N,COEF,IC,L(1),L(N),RMOM,IER)
C
      RETURN
      END
C
C
C     SUBROUTINE TO EVALUATE RATIOS OF THE MOMENTS
C     --------------------------------------------
      SUBROUTINE RATIO(NT,M,RATIO1,RATIO2)
      DIMENSION RATIO1(5),RATIO2(5)
      REAL M(5,500)
      RATIO1(NT)=M(NT,2)/M(NT,1)**2.0
      RATIO2(NT)=M(NT,1)**3.0/M(NT,3)
C     WRITE(19,*)RATIO1(NT),RATIO2(NT)
C     IF(NT.EQ.1)WRITE(19,*)
C     IF(NT.EQ.3)WRITE(19,*)
      RETURN
      END
C
C
C     SUBROUTINE FO   IMPSONS INTEGRATION
C     --------------  -------------------
      SUBROUTINE SIMP(N,H,VAL,RINT)
      DIMENSION VAL(500)
      EVEN=0
      ODD=0.
      IF(N/2*2.EQ.N)WRITE(6,*)'N NOT ODD FOR SIMPSONS'
      N1=N-1
      DO 10 I=2,N1,2
10    ODD=ODD+VAL(I)
      N2=N-2
      DO 20 I=3,N2,2
20    EVEN=EVEN+VAL(I)
      RINT=(VAL(1)+4*EVEN+2*ODD+VAL(N))*H/3.
      RETURN
```

```
      END
C
C
C     SUBROUTINE TO PRINT RESULTS
C     ---------------------------
      SUBROUTINE PRINT(Q,C,C2,RC2,CC,A,MASS,RMASS,T,CONV,M,
     #  IRUN,XPLOT,YPLOT,RATIO1,RATIO2,ALPHA,BETA,NRUN)
      DIMENSION C(5),C2(5),CC(5),A(5),F(5,500),VAL(500),SUM(5)
      DIMENSION T(5),CONV(5),COEF(500,3),RMASS(5),RC2(5)
      DIMENSION YPLOT(5,10) XPLOT(5,10),RATIO1(5),RATIO2(5)
      DIMENSION ALPHA(5),BETA(5)
      REAL L(500),MASS(5),M(5,500)
C
      WRITE(13,10)IRUN
10    FORMAT('.pa',/,20X,'RUN',I2,/,'.sk 1',/,
     #'.bx 1 15 25 35 45')
      WRITE(13,30)
30    FORMAT(18X,'FEED',5X,'TANK 1',4X,'TANK 2',/,'.bx')
C
      WRITE(13,100)Q,Q,Q,
     #RMASS(1),RMASS(2),RMASS(3), MASS(1),MASS(2),MASS(3),
     #CC(1),CC(2),CC(3),
     #C(1),C(2),C(3),RC2(1),RC2(2),RC2(3),
     #C2(1),C2(2),C2(3),
     #CONV(2),CONV(3),T(2),T(3),
     #ALPHA(2),ALPHA(3),BETA(2),BETA(3),
     #XPLOT(2,IRUN),XPLOT(3,IRUN),YPLOT(2,IRUN),YPLOT(3,IRUN),
     #M(1,1),M(2,1),M(3,1),
     #RATIO1(1),RATIO1(2),RATIO1(3),RATIO2(1),RATIO2(2),RATIO2(3)
C
100   FORMAT(2X,
     #'Q',13X,F7.3,3X,F7.3,3X,F7.3,/,2X,
     #'M by expt.',4X,F7.3,3X,F7.3,3X,F7.3,/,2X,
     #'M by M.B.',5X,F7.3,3X,F7.3,3X,F7.3,/,2X,
     #'Feaq',10X,F7.3,3X,F7.3,3X,F7.3,/,2X,
     #'Fe3+',10X,F7.3,3X,F7.3,3X,F7.3,/,2X,
     #'Fe2+',10X,F7.3,3X,F7.3,3X,F7.3,/,2X,
     #'Fe2+by M.B.',3X,F7.3,3X,F7.3,3X,F7.3,/,2X,
     #'Conversion',14X,F7.3,3X,F7.3,/,2X,
     #'τ/t*',20X,F7.3,3X,F7.3,/,2X,
     #'α',23X,F7.3,3X,F7.3,/,2X,
     #'β',23X,F7.3,3X,F7.3,/,2X,
     #'ln(Fe3+)',16X,F7.3,3X,F7.3,/,2X,
     #'A',23X,F7.3,3X,F7.3,/,2X,
     #'η',13X,F7.3,3X,F7.3,3X,F7.3,/,2X,
     #'σ/η2',10X,F7.3,3X,F7.3,3X,F7.3,/,2X,
     #'η3/ξ',10X,F7.3,3X,F7.3,3X,F7.3,/,
     #'.bx off',/,'.sk 2')
      WRITE(13,50)
50    FORMAT(
     #'A = ',
     #'ln((3ρ/φω)(Q/V)(τ/t*)η)',/,
     #'The Fe ion concentrations are in  g/l',/,
     #'Q.........................Volume flowrate l/min',/,
```

```
     #'M by expt...........Experimental mass rates g/min',/,
     #'M by M.B...Mass rates from the mass balance g/min',/,
     #'η..............................first moment    μm',/,
     #'σ.............................second moment    μm²'
     #,/,
     #'ξ..............................third moment    μm³')
C
      RETURN
      END
```

APPENDIX J. RESULTS OF PROGRAM 3

RUN 1

	FEED	TANK 1	TANK 2
Q	0.100	0.100	0.100
M by expt.	3.175	2.068	1.877
M by M.B.	3.175	2.127	2.009
Fe_{aq}	36.440	48.350	49.280
Fe^{3+}	23.800	2.830	0.480
Fe^{2+}	12.640	45.520	48.800
$Fe^{2+}$by M.B.	12.640	44.095	47.620
Conversion		33.024	5.526
τ/t^*		0.183	0.026
α		0.028	0.242
β		1.012	1.002
$\ln(Fe^{3+})$		3.925	2.151
A		-1.195	-3.339
η	192.538	159.833	157.267
σ/η^2	1.104	1.183	1.188
η^3/ξ	0.747	0.628	0.621

Q..........................*Volume flowrate* l/min

M by expt...........Experimental mass rates g/min

M by M.B...Mass rates from the mass balance g/min

η..............................first moment   μm

σ.............................second moment μm^2

ξ..............................third *moment* μm^3

The Fe ion concentrations are in g/l

$A = \ln((3\rho/\phi\omega)(Q/V)(\tau/t^*)\eta)$

RUN 2

	FEED	TANK 1	TANK 2
Q	0.150	0.150	0.150
M by expt.	3.207	2.018	1.562
M by M.B.	3.207	1.909	1.609
Fe_{aq}	35.740	48.670	49.700
Fe^{3+}	23.500	6.190	2.190
Fe^{2+}	12.240	42.480	47.510
Fe^{2+} by M.B.	12.240	38.205	44.205
Conversion		40.482	15.717
τ/t^*		0.249	0.084
α		0.021	0.079
β		1.034	1.012
$\ln(Fe^{3+})$		4.708	3.669
A		-0.484	-1.814
η	192.538	151.622	141.356
σ/η^2	1.104	1.214	1.244
η^3/ξ	0.747	0.590	0.556

Q..........................Volume flowrate l/min
M by expt...........Experimental mass rates g/min
M by M.B...Mass rates from the mass balance g/min
η..............................first moment μm
σ.............................second moment μm^2
ξ..............................third moment μm^3
The Fe ion concentrations are in g/l
$A = \ln((3\rho/\phi\omega)(Q/V)(\tau/t^*)\eta)$

RUN 3

	FEED	TANK 1	TANK 2
Q	0.200	0.200	0.200
M by expt.	3.236	1.610	1.232
M by M.B.	3.236	1.656	1.304
Fe_{aq}	35.480	45.320	47.990
Fe^{3+}	23.650	7.850	4.330
Fe^{2+}	11.830	37.470	43.660
$Fe^{2+}$by M.B.	11.830	35.530	40.810
Conversion		48.826	21.256
τ/t^{*}		0.343	0.125
α		0.015	0.056
β		1.083	1.032
$\ln(Fe^{3+})$		4.946	4.351
A		0.125	-1.184
η	192.538	143.095	130.158
σ/η^{2}	1.104	1.251	1.293
η^{3}/ξ	0.747	0.550	0.507

Q..........................Volume flowrate l/min

M by expt...........Experimental mass rates g/min

M by M.B...Mass rates from the mass balance g/min

η..............................first moment µm

σ.............................second moment μm^{2}

ξ..............................third moment μm^{3}

The Fe ion concentrations are in g/l

$A = \ln((3\rho/\phi\omega)(Q/V)(\tau/t^{*})\eta)$

RUN 4

	FEED	TANK 1	TANK 2
Q	0.250	0.250	0.250
M by expt.	3.251	1.803	1.278
M by M.B.	3.251	1.796	1.276
Fe_{aq}	37.980	44.820	48.630
Fe^{3+}	24.500	12.860	8.700
Fe^{2+}	13.480	31.960	39.930
$Fe^{2+}$by M.B.	13.480	30.940	37.180
Conversion		44.755	28.953
τ/t^*		0.294	0.182
α		0.018	0.037
β		1.055	1.048
$\ln(Fe^{3+})$		5.439	5.048
A		0.193	-0.557
η	192.538	147.152	127
σ/η^2	1.104	1.233	
η^3/ξ	0.747	0.569	[illegible]

Q..........................Volume flowrate l/min

M by expt..........Experimental mass rates g/min

M by M.B...Mass rates from the mass balance g/min

η..............................first moment μm

σ.............................second moment μm^2

ξ..............................third moment μm^3

The Fe ion concentrations are in g/l

$A = \ln((3\rho/\phi\omega)(Q/V)(\tau/t^*)\eta)$

RUN 5

	FEED	TANK 1	TANK 2
Q	0.300	0.300	0.300
M by expt.	3.117	1.602	0.971
M by M.B.	3.117	1.656	1.044
Fe_{aq}	37.720	42.990	45.760
Fe^{3+}	24.800	15.060	10.980
Fe^{2+}	12.920	27.930	34.780
Fe^{2+} by M.B.	12.920	27.530	33.650
Conversion		46.872	36.957
τ/t^*		0.319	0.260
α		0.016	0.027
β		1.068	1.092
$\ln(Fe^{3+})$		5.597	5.281
A		0.456	-0.030
η	192.538	145.017	121.052
σ/η^2	1.104	1.242	1.334
η^3/ξ	0.747	0.559	0.470

Q............................Volume flowrate l/min

M by expt...........Experimental mass rates g/min

M by M.B...Mass rates from the mass balance g/min

η..............................first moment μm

σ.............................second moment μm^2

ξ..............................third moment μm^3

The Fe ion concentrations are in g/l

$A = \ln((3\rho/\phi\omega)(Q/V)(\tau/\tau^*)\eta)$

UN 6

	FEED	TANK 1	TANK 2
Q	0.500	0.500	0.500
M by expt.	3.144	1.725	1.136
M by M.B.	3.144	1.769	1.124
Fe_{aq}	37.170	40.460	41.490
Fe^{3+}	24.600	19.100	16.520
Fe^{2+}	12.570	21.360	24.970
Fe^{2+} by M.B.	12.570	20.820	24.690
Conversion		43.734	36.461
τ/t^*		0.283	0.251
α		0.018	0.027
β		1.049	1.081
$\ln(Fe^{3+})$		5.835	5.690
A		0.846	0.466
η	192.538	148.202	123.460
σ/η^2	1.104	1.229	1.321
η^3/ξ	0.747	0.574	0.481

Q..........................Volume flowrate l/min

M by expt..........Experimental mass rates g/min

M by M.B...Mass rates from the mass balance g/min

η..............................*first moment* μm

σ..............................second moment μm^2

ξ..............................third moment μm^3

The Fe ion concentrations are in g/l

$A = \ln((3\rho/\phi\omega)(Q/V)(\tau/t^*)\eta)$

RUN 7

	FEED	TANK 1	TANK 2
Q	1.000	1.000	1.000
M by expt.	3.219	2.121	1.680
M by M.B.	3.219	2.234	1.719
Fe_{aq}	35.550	37.100	37.200
Fe^{3+}	23.700	21.730	20.700
Fe^{2+}	11.850	15.370	16.500
Fe^{2+} by M.B.	11.850	14.805	16.350
Conversion		30.600	23.053
τ/t^*		0.165	0.126
α		0.031	0.049
β		1.008	1.012
$\ln(Fe^{3+})$		5.964	5.915
A		1.002	0.562
η	192.538	162.576	144.575
σ/η^2	1.104	1.173	1.225
η^3/ξ	0.747	0.641	0.575

Q..........................Volume flowrate l/min

M by expt..........Experimental mass rates g/min

M by M.B...Mass rates from the mass balance g/min

η.............................first moment μm

σ............................second moment μm^2

ξ.............................third moment μm^3

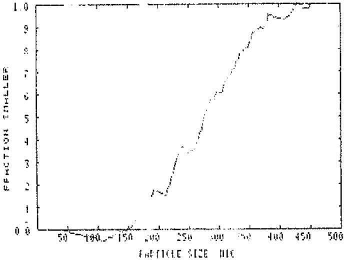
The Fe ion concentrations are in g/l

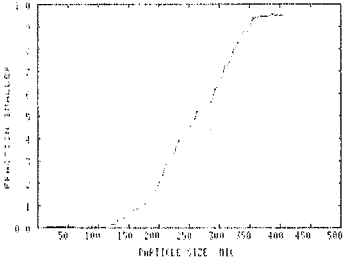
$A = \ln((3\rho/\phi\omega)(Q/V)(\tau/t^*)\eta)$

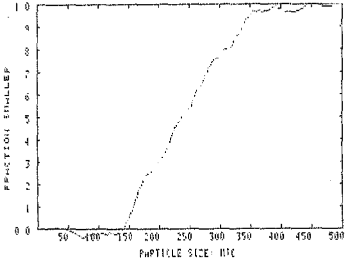
APPENDIX K. FEED SIZE DISTRIBUTION

size (µm)	f(ℓ)	size (µm)	f(ℓ)
0	0.0000000	260	0.0026285
10	0.0000000	270	0.0022759
20	0.0000000	280	0.0019724
30	0.0000000	290	0.0017038
40	0.0000000	300	0.0014621
50	0.0000000	310	0.0012451
60	0.0000000	320	0.0010509
70	0.0000000	330	0.0008737
80	0.0000000	340	0.0007077
90	0.0008497	350	0.0005536
100	0.0022647	360	0.0004204
110	0.0036269	370	0.0003146
120	0.0048798	380	0.0002345
130	0.0059155	390	0.0001759
140	0.0066797	400	0.0001335
150	0.0071453	410	0.0001002
160	0.0072755	420	0.0000709
170	0.0070908	430	0.0000455
180	0.0066836	440	0.0000258
190	0.0061433	450	0.0000125
200	0.0055452	460	0.0000043
210	0.0049642	470	0.0000001
220	0.0044398	480	0.0000000
230	0.0039569	490	0.0000000
240	0.0034899	500	0.0000000
250	0.0030383		

APPENDIX L. GRAPHS FROM THE IMAGE ANALYSER






1.0
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0.0
FRACTION SMALLER
50
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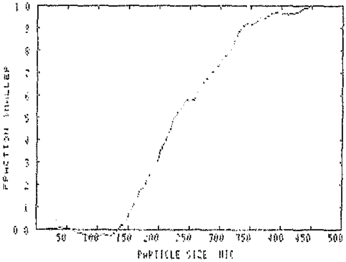
200
250
300
350
400
450
500
PARTICLE SIZE: MIC


1.0
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FRACTION SMALLER
50
100
150
200
250
300
350
400
450
500
PARTICLE SIZE: MIC

APPENDIX M. PHOTOGRAPH OF THE IRON PARTICLES

APPENDIX N. RUN WITH SAND AND WATER

The system was run using sand and water. Samples from each tank were taken then the tanks were stopped and the mass of sand and volume of water in them measured. The results are shown in the table below.

Sample	*Time (min)*	Mass(g)	Vol. (l)	Conc. (g/l)
1	10	4,09	0,825	4,96
2	15	3,95	0,815	4,85
3	20	4,10	0,820	5,00
Tank 1		4,46	0,920	4.85
Tank 2		4,77	0,930	5,13

Now

mass rate of sand into the system = 4,1 g/min

flowrate of water into the *system* = 0,81 l/*min*

therefore input concentration = 5,06 g sand/l water

Since the concentrations in the tanks and outlet streams are the same, the tanks must be well-mixed.

APPENDIX O. EXPERIMENTAL RESULTS

RUN 1

Feed Conditions

liquid flowrate = 0.100 l/min
solid Fe flowrate = 3.175 g/min
aqueous Fe concentration = 36.440 g/l
Fe^{3+} concentration = 23.800 g/l
Fe^{2+} concentration = 12.640 g/l
acid concentration = 8.700 g/l

Tank 1

Sample 1
sample volume = 0.112 l
sample mass = 2.207 g
aqueous Fe concentration = 49.000 g/l
Fe^{3+} *concentration* = 2.750 g/l
Fe^{2+} concentration = 46.250 g/l
acid concentration = 8.650 g/l

Sample 2
sample volume = 0.109 l
sample mass = 2.248 g
aqueous Fe concentration = 50.100 g/l
Fe^{3+} concentration = 2.850 g/l
Fe^{2+} concentration = 47.250 g/l
acid concentration = 8.600 g/l

Sample 3
sample volume = 0.112 l
sample mass = 2.430 g

aqueous Fe concentration = 45.950 g/l
Fe^{3+} concentration = 2.900 g/l
Fe^{2+} concentration = 43.050 g/l
acid concentration = 8.750 g/l

Tank 2

Sample 1
sample volume = 0.110 l
sample mass = 2.024 g
aqueous Fe concentration = 48.590 g/l
Fe^{3+} concentration = 0.500 g/l
Fe^{2+} concentration = 48.090 g/l
acid concentration = 8.650 g/l

Sample 2
sample volume = 0.105 l
sample mass = 2.010 g
aqueous Fe concentration = 50.220 g/l
Fe^{3+} concentration = 0.500 g/l
Fe^{2+} concentration = 49.720 g/l
acid concentration = 6.600 g/l

Sample 3
sample volume = 0.109 l
sample mass = 2.046 g
aqueous Fe concentration = 49.050 g/l
Fe^{3+} concentration = 0.450 g/l
Fe^{2+} concentration = 48.600 g/l
acid concentration = 8.600 g/l

RUN 2

Feed Conditions

liquid flowrate = 0.150 l/min
solid Fe flowrate = 3.207 g/min
aqueous Fe concentration = 35.740 g/l
Fe^{3+} concentration = 23.500 g/l
Fe^{2+} concentration = 12.240 g/l
acid concentration = 8.850 g/l

Tank 1

Sample 1
sample volume = 0.161 l
sample mass = 1.958 g
aqueous Fe concentration = 48.480 g/l
Fe^{3+} concentration = 6.050 g/l
Fe^{2+} concentration = 42.430 g/l
acid concentration = 8.850 g/l

Sample 2
sample volume = 0.165 l
sample mass = 2.061 g
aqueous Fe concentration = 49.700 g/l
Fe^{3+} concentration = 5.550 g/l
Fe^{2+} concentration = 44.150 g/l
acid concentration = 8.800 g/l

Sample 3
sample volume = 0.158 l
sample mass = 2.016 g
aqueous Fe concentration = 47.400 g/l
Fe^{3+} concentration = 6.850 g/l
Fe^{2+} concentration = 40.550 g/l
acid concentration = 8.700 g/l

Sample 4
sample volume = 0.157 l
sample mass = 2.589 g
aqueous Fe concentration = 49.080 g/l
Fe^{3+} concentration = 6.300 g/l
Fe^{2+} concentration = 42.780 g/l
acid concentration = 8.750 g/l

Tank 2

Sample 1
sample volume = 0.160 l
sample mass = 1.601 g
aqueous Fe concentration = 49.550 g/l
Fe^{3+} concentration = 2.150 g/l
Fe^{2+} concentration = 47.400 g/l
acid concentration = 8.800 g/l

Sample 2
sample volume = 0.161 l
sample mass = 1.727 g
aqueous Fe concentration = 49.690 g/l
Fe^{3+} concentration = 2.200 g/l
Fe^{2+} concentration = 47.490 g/l
acid concentration = 8.800 g/l

Sample 3
sample volume = 0.145 l
sample mass = 1.619 g
aqueous Fe concentration = 50.560 g/l
Fe^{3+} concentration = 2.150 g/l
Fe^{2+} concentration = 48.410 g/l
acid concentration = 8.850 g/l

Sample 4
sample volume = 0.159 l
sample mass = 1.653 g
aqueous Fe concentration = 48.960 g/l
Fe^{3+} concentration = 2.150 g/l

Fe^{2+} concentration = 46.810 g/l
acid concentration = 8.700 g/l

Sample 5
sample volume = 0.162 l
sample mass = 1.596 g
aqueous Fe concentration = 49.760 g/l
Fe^{3+} concentration = 2.300 g/l
Fe^{2+} concentration = 47.460 g/l
acid concentration = 8.750 g/l

Fe^{2+} concentration = 46.810 g/l
acid concentration = 8.700 g/l

Sample 5
sample volume = 0.162 l
sample mass = 1.596 g
aqueous Fe concentration = 49.760 g/l
Fe^{3+} concentration = 2.300 g/l
Fe^{2+} concentration = 47.460 g/l
acid concentration = 8.750 g/l

RUN 3

Feed Conditions

liquid flowrate = 0.200 l/min

solid Fe flowrate = 3.236 g/min

aqueous Fe concentration = 35.480 g/l

Fe^{3+} concentration = 23.650 g/l

Fe^{2+} concentration = 11.830 g/l

acid concentration = 9.800 g/l

Tank 1

Sample 1

sample volume = 0.216 l

sample mass = 1.602 g

aqueous Fe concentration = 45.610 g/l

Fe^{3+} concentration = 7.450 g/l

Fe^{2+} concentration = 38.160 g/l

acid concentration = 9.600 g/l

Sample 2

sample volume = 0.202 l

sample mass = 1.641 g

aqueous Fe concentration = 45.000 g/l

Fe^{3+} concentration = 7.750 g/l

Fe^{2+} concentration = 37.250 g/l

acid concentration = 9.700 g/l

Sample 3

sample volume = 0.208 l

sample mass = 1.700 g

aqueous Fe concentration = 45.660 g/l

Fe^{3+} concentration = 7.900 g/l

Fe^{2+} concentration = 37.760 g/l

acid concentration = 9.750 g/l

Sample 4

sample volume = 0.212 l
sample mass = 1.801 g
aqueous Fe concentration = 45.020 g/l
Fe^{3+} concentration = 8.300 g/l
Fe^{2+} concentration = 36.720 g/l
acid concentration = 9.750 g/l

Tank 2

Sample 1

sample volume = 0.215 l
sample mass = 1.352 g
aqueous Fe concentration = 48.040 g/l
Fe^{3+} concentration = 4.000 g/l
Fe^{2+} concentration = 44.040 g/l
acid concentration = 9.700 g/l

Sample 2

sample volume = 0.210 l
sample mass = 1.335 g
aqueous Fe concentration = 48.040 g/l
Fe^{3+} concentration = 4.400 g/l
Fe^{2+} concentration = 43.640 g/l
acid concentration = 9.700 g/l

Sample 3

sample volume = 0.208 l
sample mass = 1.282 g
aqueous Fe concentration = 47.980 g/l
Fe^{3+} concentration = 4.500 g/l
Fe^{2+} concentration = 43.480 g/l
acid concentration = 9.750 g/l

Sample 4

sample volume = 0.207 l
sample mass = 1.269 g
aqueous Fe concentration = 47.920 g/l
Fe^{3+} concentration = 4.500 g/l

Fe^{2+} concentration = 43,420 g/l
acid concentration = 9,700 g/l

Sample 5 sample volume = 0,213 l
sample mass = 1,251 g
aqueous Fe concentration = 47,970 g/l
Fe^{3+} concentration = 4,250 g/l
Fe^{2+} concentration = 43,720 g/l
acid concentration = 9,650 g/l

RUN 4

Feed Conditions

liquid flowrate = 0.250 l/min
solid Fe flowrate = 3.251 g/min
aqueous Fe concentration = 37.980 g/l
Fe^{3+} concentration = 24.500 g/l
Fe^{2+} concentration = 13.480 g/l
acid concentration = 8.700 g/l

Tank 1

Sample 1
sample volume = 0.262 l
sample mass = 1.835 g
aqueous Fe concentration = 44.700 g/l
Fe^{3+} concentration = 13.000 g/l
Fe^{2+} concentration = 31.700 g/l
acid concentration = 8.700 g/l

Sample 2
sample volume = 0.260 l
sample mass = 1.970 g
aqueous Fe concentration = 44.610 g/l
Fe^{3+} concentration = 13.650 g/l
Fe^{2+} concentration = 30.960 g/l
acid concentration = 8.750 g/l

Sample 3
sample volume = 0.261 l
sample mass = 1.874 g
aqueous Fe concentration = 44.970 g/l
Fe^{3+} concentration = 12.350 g/l
Fe^{2+} concentration = 32.620 g/l
acid concentration = 8.600 g/l

Sample 4
sample volume = 0.246 l
sample mass = 1.743 g
aqueous Fe concentration = 45.020 g/l
Fe^{3+} concentration = 12.450 g/l
Fe^{2+} concentration = 32.570 g/l
acid concentration = 6.650 g/l

Tank 2

Sample 1
sample volume = 0.251 l
sample mass = 1.310 g
aqueous Fe concentration = 46.610 g/l
Fe^{3+} concentration = 8.550 g/l
Fe^{2+} concentration = 38.060 g/l
acid concentration = 8.700 g/l

Sample 2
sample volume = 0.255 l
sample mass = 1.267 g
aqueous Fe concentration = 46.490 g/l
Fe^{3+} concentration = 8.550 g/l
Fe^{2+} concentration = 37.940 g/l
acid concentration = 8.500 g/l

Sample 3
sample volume = 0.257 l
sample mass = 1.330 g
aqueous Fe concentration = 47.320 g/l
Fe^{3+} concentration = 9.000 g/l
Fe^{2+} concentration = 38.320 g/l
acid concentration = 8.650 g/l

Sample 4
sample volume = 0.261 l
sample mass = 1 359 g
aqueous Fe concentration = 55.650 g/l
Fe^{3+} concentration = 9.000 g/l

Fe^{2+} concentration = 46.650 g/l
acid concentration = 8.700 g/l

Sample 5
sample volume = 0.254 l
sample mass = 1.267 g
aqueous Fe concentration = 47.090 g/l
Fe^{3+} concentration = 8.400 g/l
Fe^{2+} concentration = 38.690 g/l
acid concentration = 8.700 g/l

RUN 5

Feed Conditions

liquid flowrate = 0.300 l/min

solid Fe flowrate = 3.117 g/min

aqueous Fe concentration = 37.720 g/l

Fe^{3+} concentration = 24.800 g/l

Fe^{2+} concentration = 12.920 g/l

acid concentration = 9.250 g/l

Tank 1

Sample 1
sample volume = 0.296 l
sample mass = 1.702 g
aqueous Fe concentration = 41.150 g/l
Fe^{3+} concentration = 15.700 g/l
Fe^{2+} concentration = 25.450 g/l
acid concentration = 9.100 g/l

Sample 2
sample volume = 0.286 l
sample mass = 1.468 g
aqueous Fe concentration = 43.870 g/l
Fe^{3+} concentration = 14.300 g/l
Fe^{2+} concentration = 29.570 g/l
acid concentration = 9.150 g/l

Sample 3
sample volume = 0.328 l
sample mass = 1.622 g
aqueous Fe concentration = 43.860 g/l
Fe^{3+} concentration = 14.750 g/l
Fe^{2+} concentration = 29.110 g/l
acid concentration = 9.150 g/l

Sample 4
sample volume = 0.290 l
sample mass = 1.617 g
aqueous Fe concentration = 43.100 g/l
Fe^{3+} concentration = 15.500 g/l
Fe^{2+} concentration = 27.600 g/l
acid concentration = 9.100 g/l

Tank 2

Sample 1
sample volume = 0.306 l
sample mass = 1.066 g
aqueous Fe concentration = 45.340 g/l
Fe^{3+} concentration = 11.250 g/l
Fe^{2+} concentration = 34.090 g/l
acid concentration = 9.200 g/l

Sample 2
sample volume = 0.320 l
sample mass = 0.983 g
aqueous Fe concentration = 45.860 g/l
Fe^{3+} concentration = 10.600 g/l
Fe^{2+} concentration = 35.260 g/l
acid concentration = 9.150 g/l

Sample 3
sample volume = 0.300 l
sample mass = 1.037 g
aqueous Fe concentration = 45.490 g/l
Fe^{3+} concentration = 11.300 g/l
Fe^{2+} concentration = 34.190 g/l
acid concentration = 9.150 g/l

Sample 4
sample volume = 0.335 l
sample mass = 0.970 g
aqueous Fe concentration = 46.380 g/l
Fe^{3+} concentration = 10.450 g/l

Fe^{2+} concentration = 35.930 g/l
acid concentration = 9.100 g/l

Sample 5 sample volume = 0.328 l
sample mass = 1.088 g
aqueous Fe concentration = 45.710 g/l
Fe^{3+} concentration = 11.300 g/l
Fe^{2+} concentration = 34.410 g/l
acid concentration = 9.050 g/l

RUN 6

Feed Conditions

liquid flowrate = 0.500 l/min
solid Fe flowrate = 3.144 g/min
aqueous Fe concentration = 37.170 g/l
Fe^{3+} concentration = 24.600 g/l
Fe^{2+} concentration = 12.570 g/l
acid concentration = 9.400 g/l

Tank 1

Sample 1
sample volume = 0.252 l
sample mass = 0.831 g
aqueous Fe concentration = 40.640 g/l
Fe^{3+} concentration = 18.650 g/l
Fe^{2+} concentration = 21.990 g/l
acid concentration = 9.450 g/l

Sample 2
sample volume = 0.256 l
sample mass = 0.867 g
aqueous Fe concentration = 40.450 g/l
Fe^{3+} concentration = 19.000 g/l
Fe^{2+} concentration = 21.450 g/l
acid concentration = 9.400 g/l

Sample 3
sample volume = 0.259 l
sample mass = 0.941 g
aqueous Fe concentration = 40.210 g/l
Fe^{3+} concentration = 19.500 g/l
Fe^{2+} concentration = 20.710 g/l
acid concentration = 9.350 g/l

Sample 4
sample volume = 0.255 l
sample mass = 0.887 g
aqueous Fe concentration = 40.540 g/l
Fe^{3+} concentration = 19.250 g/l
Fe^{2+} concentration = 21.290 g/l
acid concentration = 9.300 g/l

Tank 2

Sample 1
sample volume = 0.247 l
sample mass = 0.518 g
aqueous Fe concentration = 41.900 g/l
Fe^{3+} concentration = 16.200 g/l
Fe^{2+} concentration = 25.700 g/l
acid concentration = 9.300 g/l

Sample 2
sample volume = 0.257 l
sample mass = 0.624 g
aqueous Fe concentration = 41.440 g/l
Fe^{3+} concentration = 17.000 g/l
Fe^{2+} concentration = 24.440 g/l
acid concentration = 9.350 g/l

Sample 3
sample volume = 0.255 l
sample mass = 0.583 g
aqueous Fe concentration = 41.130 g/l
Fe^{3+} concentration = 16.350 g/l
Fe^{2+} concentration = 24.780 g/l
acid concentration = 9.250 g/l

RUN 7

Feed Conditions

liquid flowrate = 1.000 l/min
solid Fe flowrate = 3.219 g/min
aqueous Fe concentration = 35.550 g/l
Fe^{3+} concentration = 23.700 g/l
Fe^{2+} concentration = 11.850 g/l
acid concentration = 9.500 g/l

Tank 1

Sample 1
sample volume = 0.260 l
sample mass = 0.515 g
aqueous Fe concentration = 37.620 g/l
Fe^{3+} concentration = 21.800 g/l
Fe^{2+} concentration = 15.820 g/l
acid concentration = 9.500 g/l

Sample 2
sample volume = 0.245 l
sample mass = 0.588 g
aqueous Fe concentration = 36.370 g/l
Fe^{3+} concentration = 21.750 g/l
Fe^{2+} concentration = 14.620 g/l
acid concentration = 9.550 g/l

Sample 3
sample volume = 0.263 l
sample mass = 0.526 g
aqueous Fe concentration = 37.330 g/l
Fe^{3+} concentration = 21.650 g/l
Fe^{2+} concentration = 15.680 g/l
acid concentration = 9.500 g/l

Tank 2

Sample 1
sample volume ≈ 0.257 l
sample mass ≈ 0.438 g
aqueous Fe concentration ≈ 37.000 g/l
Fe^{3+} concentration ≈ 20.800 g/l
Fe^{2+} concentration ≈ 16.200 g/l
acid concentration ≈ 9.450 g/l

Sample 2
sample volume ≈ 0.259 l
sample mass ≈ 0.429 g
aqueous Fe concentration ≈ 37.400 g/l
Fe^{3+} concentration ≈ 20.600 g/l
Fe^{2+} concentration ≈ 16.800 g/l
acid concentration ≈ 9.550 g/l

APPENDIX P. NOMENCLATURE

Symbol Quantity

$a(\ell)$ = exposed surface area of particle of size ℓ
C_i = *fractional conversion in tank i*
E = potential
E° = standard equilibrium potential
F = Faraday's constant
$f_i(\ell)d\ell$ = fraction of particles in tank i with sizes between ℓ and $\ell+d\ell$
i = current density
i_0 = exchange current density
i_0^* = exchange current density independent of concentration
k_i = rate constant in tank i
ℓ = *characteristic particle size*
= $[m(\ell)/\rho]^{1/3}$
$m(\ell)$ = mass of single particle of size ℓ
n = no. of tanks
N_i = no. of particles in tank i per unit volume of solution
Q = volumetric flowrate of solution
R = gas constant
T_i = τ_i/t^*
t = residence time of particle in contact with the solution
t_i^* = $-\ell_{i-1}/u_i$
u_i = particle growth rate in tank i
= $d\ell/dt$
V_i = volume of tank i
z = no. of electrons transferred

ρ = density of particle

ϕ = shape factor

= $a(\ell)/\ell^2$

ψ_i = quantity dependant on chemical and physical properties of the solution environment in tank i

τ_i = V_i/Q

ω = moleculer weight of dissolving species

η_i = $\int_0^\infty \ell \, f_i(\ell) d\ell$

σ_i = $\int_0^\infty \ell^2 f_i(\ell) d\ell$

ξ_i = $\int_0^\infty \ell^3 f_i(\ell) d\ell$

Author Comninos Peter Harry
Name of thesis A Number Balance Approach To The Modelling Of The Leaching Of Particles In Multiple Well-mixed Tanks. 1985