DOE/PC/94217--T7
DOE/MST-9506

**A STUDY OF
MULTISTAGE/MULTIFUNCTION COLUMN
FOR FINE PARTICLE SEPARATION**

QUARTERLY TECHNICAL PROGRESS REPORT
(January 1, 1996 - March 31, 1996)

RECEIVED
AUG 05 1996
OSTI

Principal Investigator: Dr. Shiao-Hung Chiang
Project Officer: Dr. Ralph W. Lai

Work Performed under DOE Contract No. DE-PG22-94PC94217

(Award: \$200,000; Duration: July 8, 1994 - July 7, 1997)

Submitted to

**U.S. Department of Energy
Pittsburgh Energy Technology Center
Pittsburgh, PA 15236-0940**

by

**Chemical and Petroleum Engineering Department
University of Pittsburgh
Pittsburgh, PA 15261**

April 20, 1996

RECEIVED
USDOE/PETC
36 APR 17 AM 10:19
ACQUISITION & ASSISTANCE DIV.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

**Portions of this document may be illegible
in electronic image products. Images are
produced from the best available original
document.**

1.0 INTRODUCTION

The overall objective of the proposed research program is to explore the potential application of a new invention involving a multistage column equipped with vortex-inducing loop-flow contactors (hereafter referred to as **the multistage column**) for fine coal cleaning process. The research work will identify the design parameters and their effects on the performance of the separation process. The results of this study will provide an engineering basis for further development of this technology in coal cleaning and in the general areas of fluid/particle separation.

In the last quarter, we investigated the mixing and loop flow (circulation) behaviors around the contactors. In this quarter, the fine coal beneficiation tests were carried out in the multistage column and conventional column. Table 1.1, the project schedule, shows work accomplished to date.

Table 1.1 Project Schedule

Tasks	1994			1995			1996			1997			
	7	9	12	3	6	9	12	3	6	9	12	3	6
1. Project Planning	■												
2. Equipment		■	■	■									
3. Hydrodynamic Tests				■	■	■	■						
4. Separation Tests							■	■	■	■	■	■	■
5. Conventional Column Tests				■	■	■	■						
6. Data Analysis				■	■	■	■	■	■	■	■	■	■
7. Reports	□	□	□	■	□	□	□	■	□	□	□	□	●

Notes: □ Quarterly Technical Progress Report; ■ Annual Report; ● Final Report.

2.0 TECHNICAL PROGRESS

2.1 Task 1: Project Planning

This task was completed in September 1994^[1].

2.2 Task 2: Equipment Design and Construction

Task 2 was completed in March 1995^[2].

2.3 Task 3: Hydrodynamic Tests

The objective of this task is to pursue a basic understanding of the hydrodynamic behavior and to characterize the flow and mixing conditions in the multistage separation column.

In the last quarter, we have completed the following subtasks:

1. The investigation of mixing behavior in the multistage column and conventional column, which include:
 - a. Setting up and calibration of the conductivity meter and auxiliary devices;
 - b. Measurements of mixing time under various operating conditions.
2. The investigation of circulation flow characteristics around the contactor, which include:
 - a. Definition of the circulation velocity and development of a measurement procedure;
 - b. Measurements of the circulation velocity around the contactors.

All the subtasks in Task 3 were completed in March 1996^[3-5].

2.4 Task 4: Separation Tests

Task 4 has been initiated in January 1996. The goal of this task is to conduct the investigation on fine coal beneficiation and evaluate the effectiveness of the multistage flotation

column. In addition, the potential application of the multistage column to other separation processes will be explored.

In this quarter, to verify the advantages of using the multistage column in the fine coal flotation process, a series of coal cleaning tests were carried out to compare the performance of the multistage column with that of the conventional column.

2.4.1 Experimental Procedure and Operating Conditions

In the study of coal cleaning process, the heating value recovery (*HVR*) and the impurity rejection were the criteria used in the evaluation of column performance. The *HVR* and impurity rejection (including pyrite rejection and ash rejection) are defined by Equations (2.1) and (2.2), respectively, as follows:

$$HVR = \frac{(BTU)_{\text{recovered}}}{(BTU)_{\text{total}}} = \frac{\sum_{i=1}^I W_i (BTU)_i}{(BTU)_{\text{total}}} \quad (2.1)$$

where W_i is the weight of coal recovered in the i th collection; $(BTU)_i$ is the *BTU* per unit weight in the i th collection; $(BTU)_{\text{total}}$ is the total *BTU* in the feed coal.

$$(\text{rejection})_{\text{impurity}} = 1 - \frac{\sum_{i=1}^I W_i (IP)_i}{(IP)_{\text{total}}} \quad (2.2)$$

where $(IP)_i$ is the impurity content in the i th collection; $(IP)_{\text{total}}$ is the total impurity content in the feed coal.

The Upper Freeport coal from Indiana County, Pennsylvania was selected as the model coal for this study. The coal was crushed and screened to collect the 100×325 mesh size fraction as the feed coal sample. The mean size of the feed coal was 45-150 μm . The feed coal sample contained 26.4% ash and 2.95% sulfur (2.4% pyritic sulfur).

In each batch test, 300 grams of coal were premixed with 500 ml tap water to prepare the slurry feed. Various dosages of frother, 4-methyl-2-pentanol (MIBC), were added to the liquid phase and premixed for 5 minutes. The typical solid concentration in the column was approximately 3%. Clean coal samples were collected at predetermined time intervals (usually 0.5, 1, 2, 4, 8, 16, 32 minutes) until the froth was depleted. The beneficiation tests were also conducted in the conventional column under the same operating conditions.

2.4.2 Heating Value Recovery

The *HVR* versus flotation time is plotted in Figure 2.1. It can be seen from the plot that the conventional flotation column, recovered only about half of the heating value compared to the multistage column. In the multistage column, during the first 4 minutes, more than 85% of the heating value was already recovered. This confirmed that the better kinetic performance in the multistage column was mainly due to the arrangement of draft tubes and attached vortex inducing plates.

2.4.3 Impurity Rejection

In addition to *HVR*, the other key performance criterion of a fine coal flotation process is pyrite rejection. The current interest centers on pyrite rejection because the sulfur components in coal are the root cause of SO_2 emission in coal-fired power plants. Figure 2.2 presents a comparison of pyritic sulfur rejection between the multistage column and the conventional column. It can be seen from Figure 2.2 that the multistage column achieves a much greater pyrite rejection with higher *HVR* than the conventional column.

Figure 2.1 Comparison of HVR between Multistage Column and Conventional Column

Note: SGV- Superficial Gas Velocity.

Figure 2.2 Comparison of Pyrite Rejection between Multistage Column and Conventional Column

Further, it is shown that even when the *HVR* in the multistage column is as high as 95%, the pyrite rejection still remains above 70%. This indicates that a very significant improvement in the *HVR* can be achieved in the multistage column without seriously affecting the pyritic sulfur rejection. In other words, the multistage column can provide a good product quality over a considerably wider

operating range than the conventional column. This finding is in good agreement with the results documented in our previous work^[6-8]. It is important to note that such a favorable performance is essential for the operation of a commercial flotation column.

2.4.3 Role of the Frother Dosage

Since the addition of frother could alter the column hydrodynamic state and, in turn, the column beneficiation behavior, the effect of frother dosage on the column separation characteristics was also studied in the multistage and conventional columns. The data for the effect of MIBC dosage (*C*) obtained from the conventional column and multistage column are plotted in Figures 2.3 and 2.4, respectively. As shown in Figure 2.3, in the conventional column the pyrite rejection and the *HVR* are very sensitive to the change in the frother dosage. Although the *HVR* increases significantly as the MIBC dosage increases, the pyrite rejection decreases sharply. This is attributed to the deficient control of impurity entrainment in the conventional column. On the other hand, the pyrite rejection is less sensitive to the variation in MIBC dosage in the multistage column. The results (see Figure 2.4) indicate that using a low MIBC dosage of 15 ppm is sufficient to obtain a high pyrite rejection. This is because in the conventional column the bubble sizes are large and not uniformly distributed^[4-5] as observed in the hydrodynamic tests. In the multistage column, the bubble sizes are uniform and relatively small, resulting in a large interfacial area for bubble-particle contacts.

Figure 2.3 The Effects of Frother Dosage on Pyrite Rejection (Conventional Column)

Figure 2.4 The Effects of Frother Dosage on Pyrite Rejection (Multistage Column)

2.4.4 Fine Coal Beneficiation Tests Under Various Operating Conditions

To identify the effect of operating conditions on the characteristics of fine coal flotation in the multistage column, additional tests were carried out. The results of these tests are presented in

Table 2.1. As noted in Table 2.1, the results indicate that the multistage column demonstrates a better operating flexibility.

Table 2.1 Performances of Multistage Fine Coal Flotation

Test	1	2	3	4
MIBC, ppm	15	15	20	20
N, Hz	30	30	24	15
SVG, m/s	0.01	0.006	0.006	0.006
Maximum <i>HVR</i> (%)	95.1	93.7	96.5	89.4
Maximum Pyrite Rejection (%)	67.5	73.5	70.3	76.3
Maximum Ash Rejection (%)	78.6	75.6	77.8	74.3
<i>HVR</i> at 4 minutes (%)	87.9	86.5	84.4	74.2

2.5 Task 5: Conventional Column Tests

In the last quarter, we completed the investigation on the liquid mixing and circulation behavior in the conventional flotation column. The results were reported in the last quarterly report^[5]. In this quarter, experiments similar to those for Task 4 were carried out to study the fine coal beneficiation behavior in the conventional column for comparison purposes. The results have been discussed in Section 2.4.

2.6 Task 6: Data Analysis

In the past several quarters, data for the gas holdup, bubble size and specific interfacial area^[4] were analyzed in terms of three key operating parameters: SGV, agitation speed (N) and C. Empirical correlations were established in terms of three second-order polynomial equations^[4] representing the experimental data.

In this quarter, an attempt was made to analyze the kinetic data obtained from the multistage column using a flotation model based on the unified proportionality equation^[9-11].

2.6.1 Basic Principle of the Model Equation

The unified proportionality equation is a multiscale, multidimensional equation. The equation not only embraces both linear and logarithmic scales, but also includes a multilogarithmic dimension. The equation has shown to be useful for modeling experimental data when dealing with two associated variables ^[9-11]. In this study, we performed the modeling of flotation kinetic data using three forms of equations derived from the general unified proportionality equation. The kinetic data on fine coal flotation in the multistage column were used for statistical discrimination of the models. Model equation parameters were obtained using Microsoft Excel (version 5.0). Statistical parameters were used for comparison of model applicability and identification of the most desirable model. Graphs for expressing the proportionality of each model are discussed below.

2.6.1.1 General Equation

The unified proportionality equation is given as:

$$d(q^m Y) = K d(q^n X) \quad (2.3)$$

where q is a notation of logarithm, K is a proportionality constant, and m and n are logarithmic dimensions of Y and X . The effective $q^m Y$ and $q^n X$ always refer to the distance between $q^m Y$ or $q^n X$ and its respective asymptote.

In Equation (2.3), m and n can be integers 0, 1, 2, 3,... etc. When using the different values of m and n , Equation (2.3) can be written as the following expressions:

$$dY = K dX \quad (2.3.a)$$

$$d(\log Y) = K dX \quad (2.3.b)$$

$$d(\log Y) = K d(\log X) \quad (2.3.c)$$

$$d[\log(\log Y)] = K \frac{dX}{X} \quad (2.3.d)$$

Equation (2.3.a) does not have an asymptote associated with either Y or X . Thus, this equation can not be used to model the flotation kinetic data. The other three model equations need to incorporate the recovery asymptote (Y_i) as reference point for accounting the effective recovery (Y), as will be discussed in the next section.

2.6.1.2 Flotation Recovery Models - phenomena with ($Y_i > Y$)

In flotation, we are most interested in finding how much floatable material can be recovered or knowing what the recovery profile looks like as the flotation progresses in time. Because the profile of cumulative recovery tends to approach an asymptote, it is imperative that the asymptote be identified first and be used as a key reference in process analysis of effective recovery.

The integral forms of the flotation recovery models, Equations (2.3.b) through (2.3.d), are:

$$\log \frac{1}{(Y_i - Y)} = KX + \log A \quad (\text{Model 1}) \quad (2.4)$$

$$\log \frac{1}{(Y_i - Y)} = K \log X + \log A \quad (\text{Model 2}) \quad (2.5)$$

$$\log \frac{1}{(qY_i - qY)} = K \log X + \log A \quad (\text{Model 3}) \quad (2.6)$$

In the above equations, the term $1/(Y_i - Y)$ and $1/(qY_i - qY)$ are known as the *proportionality factors*. In Equations (2.4) and (2.5), the *log* implies the intention of plotting the proportionality factor $1/(Y_i - Y)$ in a logarithmic scale. In Equation (2.6), qY_i and qY emphasize the need to calculate the values of $\log(Y_i)$ and $\log(Y)$; and the *log* implies the intention of plotting the proportionality factor $1/(qY_i - qY)$ in a logarithmic scale. According to Equation (2.4), $1/(Y_i - Y)$ versus X can be plotted in a semi-log graph to obtain a straight line if the proportionality exists. Similarly, according to Equations (2.5) and (2.6), $1/(Y_i - Y)$ versus X or $1/(qY_i - qY)$ versus X can be plotted in a log-log graph to obtain a straight line if the proportionality exists. Microsoft Excel was used to identify the asymptote Y_i and parameters A and K in the above model equations.

2.6.2 Data Analysis Procedure

Microsoft Excel was used for data analysis and graphing. Using an object oriented interactive approach, the asymptote Y_i was first sought. For each model, the K , A , and unique Y_i were determined simultaneously for Equations (2.4), (2.5), and (2.6) using the trendline (or so-called regression) analysis. The targeted objective was to identify the unique asymptote Y_i for the best fit of the trendline or the maximum of the *coefficient of determination* (R^2). The value of R^2 can be defined as follows:

$$R^2 = 1 - \frac{SSE}{SSTO} = 1 - \frac{\sum (Y_i - \hat{Y}_i)^2}{\sum (Y_i - \bar{Y})^2} \quad (2.7)$$

where, *SSE* is the error sum of squares, which is used to measure the variation in the observations Y_i when a regression model utilizing the independent variable X is employed; *SSTO* is a measure of the uncertainty in predicting Y when the independent variable X is not considered.

The flotation data obtained from the multistage column and the calculated proportionality factors are listed in Table 2.3. Based on regression analysis, the model parameters were determined and presented in Table 2.4. Using the model parameters in Table 2.4, the theoretical Y values were calculated for each model (see Table 2.5). The mean square errors (*MSE*) were then calculated for the differences between the theoretical and experimental data, and used as indicators to compare the superiority of the models (see Table 2.6). The *MSE* is defined by the following expression:

$$MSE = \frac{\sum_{j=1}^n (Y_j - \hat{Y}_j)^2}{n - m} \quad (2-8)$$

Table 2.3 Flotation Kinetic Data Obtained From Multistage Column

X	Y	Y_i		
		96.501	98.4	97.3
Time, min	HVR, %	$1/(Y_i - Y)$	$1/(Y_i - Y)$	$1/(qY_i - qY)$
0.5	19.0	0.012903	0.012594	1.409723
1	37.8	0.017035	0.016502	2.435336
2	61.3	0.028408	0.026954	4.983744
4	84.4	0.082638	0.071429	16.18899
8	93.2	0.302939	0.192308	53.48472
16	96.1	2.493766	0.434783	185.5476
32	96.5	1000.000	0.526316	278.899

Table 2.4 Model Parameters

Model	1	2	3
Y_i	96.501	98.4	97.3
R^2	0.9951	0.9708	0.9835
K	0.15076	1.0153	1.3861
A	0.014	0.0191	2.7208

Table 2.5 Comparison of Calculated HVR with Experimental HVR

Time, min	Experimental HVR, %	Model 1 %	Model 2 %	Model 3 %
0.5	19.0	36.4539	7.42844	10.65300
1	37.8	46.02184	46.04398	41.74206
2	61.3	60.82697	72.49814	70.38714
4	84.4	78.68409	85.58569	85.96311
8	93.2	92.05681	92.06044	92.79591
16	96.1	96.22449	95.26366	95.55148
32	96.5	96.49993	96.84837	96.62727

2.6.3 Discussion and Remark

For a better discrimination of the models, we rely on a quantitative statistical parameter for comparison. Table 2.6 gives a comparison of the *MSE* for the three flotation kinetic models. It is clearly shown that Model 3 has the lowest MSE, Model 1 has moderate MSE, and Model 2 has the

highest *MSE*. It has been found in other studies that in general, the *MSE* of Model 1 is one or two orders of magnitude higher than that of Model 3^[11]. Based on the *MSE* comparison, it can be seen that Model 3 (Equation (2.6)) is the most applicable model for batch flotation data.

Table 2.6 Statistical Discrimination Analysis

Model	Square Error		
	1	2	3
	304.6387	698.4624	69.67239
	67.59872	67.96319	15.53984
	0.223760	125.3984	82.57603
	32.67166	1.405868	2.443318
	1.306891	1.298605	0.163293
	0.015497	0.699470	0.300869
	4.96E-09	0.121362	0.016199
SUM=	406.4552	895.3493	170.7119
<i>MSE</i> =	58.06503	127.907	24.38742

Figures 2.5 through 2.7 depict a comparison of the experimental data with the calculated data based on the three models. Figure 2.5 gives a proportionality plot for Model 1, where the proportionality factor $1/(Y_i - Y)$ is plotted against time in a semi-log graph. Figure 2.6 gives a proportionality plot for Model 2, where the proportionality factor $1/(Y_i - Y)$ is plotted against time in a log-log graph. Figure 2.7 is a proportionality plot for Model 3, where the proportionality factor $1/(qY_i - qY)$ is plotted against time in a log-log graph. In the above figures, all trendlines are drawn to give unique Y_i using the Excel. A comparison of Figures 2.5, 2.6, and 2.7 reveals that the data points and the trendlines are most matched by Model 3.

Figure 2.5 Proportionality Plot for Model 1

Figure 2.6 Proportionality Plot for Model 2

Figure 2.7 Proportionality Plot for Model 3

In Table 2.4, the R^2 values range from 0.9708 to 0.9951. These R^2 values for all models are high, indicating that all models can potentially be considered as a feasible candidate model. However, in reality Models 1 and 2 are not desirable, as verified quantitatively with *MSE* values. In his analysis of *MSE* for batch flotation data, Mazumdar ^[12] also confirmed that Model 3 is best and Model 1 is worst among the models tested.

From the above discussion, it can be concluded that the relationship of recovery with time for the multistage column flotation is not linear. Instead, their relationship is changing in a logarithmic scale and their relative change is varying in a fashion of fractional change rather than simple linear change. For this reason, the flotation kinetics can best be described by a high dimensional subset of the unified proportionality equation, which may be written as:

$$\frac{1}{\log Y_i - \log Y} = 2.7208 X^{1.3861} \quad (2.9)$$

where, X is flotation time, min.

2.7 Summary

In the past several quarters, the mathematical expressions developed using the multiparameter regression method provided adequate correlations for the column hydrodynamic data. In this quarter, we conducted the investigation of fine coal flotation in the multistage column in terms of *HVR* and impurity rejection. Additionally, we identified the applicability of three flotation kinetic models for multistage column kinetic data correlation. In conclusion, the following points are noted:

1. The experimental investigation of the fine coal flotation in the multistage column shows that the design concept of the multistage column equipped with vortex-inducing contactors is expected to result in an improved performance compared to the conventional column.
2. The superficial gas velocity and the agitation speed are two operating parameters affecting the *HVR* and impurity rejection.
3. When the MIBC dosage is greater than 15 ppm, a further increase in the MIBC dosage does not play a significant role in affecting the column operating performance.
4. The fine coal beneficiation behavior of the multistage column appears to be less sensitive to the variation of MIBC dosages than that of the conventional column.
5. Model 3, i.e. Equation (2.6), is the most applicable model for describing the batch flotation data obtained from the multistage column.

3.0 WORK FORECAST

In the next quarter, separation tests will be conducted as part of work under Task 4 to study the application of the multistage flotation column to other separation processes and to demonstrate its multifunction capability. Work will include the following aspects:

- Waste water treatment;
- Recycled paper pulp de-inking;
- Removing metal oxide precipitates from decon water.

4.0 REFERENCES

1. *"A Study of Multistage/Multifunction Column for Fine Particle Separation"*, Quarterly Technical Progress Report, (July 8, 1994 - September 30, 1994), Chemical & Petroleum Engineering Department, University of Pittsburgh, Submitted to U.S. Department of Energy, PETC, January 20, 1995.
2. *"A Study of Multistage/Multifunction Column for Fine Particle Separation"*, Quarterly Technical Progress Report, (October 1 - December 31, 1994), Chemical & Petroleum Engineering Department, University of Pittsburgh, Submitted to U.S. Department of Energy, PETC, February 10, 1995.
3. *"A Study of Multistage/Multifunction Column for Fine Particle Separation"*, Quarterly Technical Progress Report, (April 1, 1995 - June 30, 1995), Chemical & Petroleum

Engineering Department, University of Pittsburgh, Submitted to U.S. Department of Energy, PETC, July 20, 1995.

4. "A Study of Multistage/Multifunction Column for Fine Particle Separation", Quarterly Technical Progress Report, (July 1, 1995 - September 30, 1995), Chemical & Petroleum Engineering Department, University of Pittsburgh, Submitted to U.S. Department of Energy, PETC, October 20, 1995.
5. "A Study of Multistage/Multifunction Column for Fine Particle Separation", Quarterly Technical Progress Report, (October 1, 1995 - December 31, 1995), Chemical & Petroleum Engineering Department, University of Pittsburgh, Submitted to U.S. Department of Energy, PETC, January 20, 1995.
6. D.X. He and S.H. Chiang, "Further Investigation of the Novel Multistage Agitated Column for Fine Coal Beneficiation", Fluid/Particle Separation Journal, 7(4), Dec. 1994, p.137.
7. D.X. He and S.H. Chiang, "An Investigation of Novel Multistage Agitated Column for Precombustion Coal Cleaning", Proceedings of the Third Asian-Pacific International Symposium on Combustion and Energy Utilization, Hong Kong, Dec. 11-15, 1995, p.775.
8. R. W. Lai, R. A. Patton, D. X. He, T. Joyce, and S. H. Chiang, "Initial Testing of a Dynamic Column for Fine Coal Flotation," Proceed. 12th Ann. Int. Pitt. Coal Conf., Sept.11-15, 1995, p. 851.
9. R. W. Lai, "How to Obtain More Information Out of Flotation Kinetics Data," Trans. AIME, Vol. 272, 1982, p. 1989.

10. R. W. Lai, M. W. Lai, J. A. Musser, and P. C. Rehmet, *The Modern Proportionality Law for Science, Medicine, and Engineering Applications*, The Toshi Company, Pittsburgh, PA, 1995, p. 384.
11. R. W. Lai, D. X. He, and S.H. Chiang, "Flotation Models Based on The Unified Proportionality Equation," to be presented at *1996 AFS Annual Technical Conference*, Valley Forge, Pennsylvania, April 21-24, 1996.
12. M. Mazumdar, "Statistical Discrimination of Flotation Models Based on Batch Flotation Data," *Int. J. Miner. Process.*, Vol.42, 1994, p. 53.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.