DOE/MC/11089--3203

DE93 000242

Pressurized Fluidized-Bed Hydroretorting of Eastern Oil Shales Oil Dedusting Subtask 3.4: Electroseparation of Fines From Shale Oil

Topical Report

F.S. Lau
D. Gidaspow
U. Jayaswal
D.T. Wasan

November 1991

Work Performed Under Contract No.: DE-AC21-87MC11089

For
U.S. Department of Energy
Office of Fossil Energy
Morgantown Energy Technology Center
Morgantown, West Virginia

By
Institute of Gas Technology
Chicago, Illinois
and
Illinois Institute of Technology
Chicago, Illinois

DISCLAIMER

EXECUTIVE SUMMARY

This Topical Report on "Shale Oil Dedusting" presents the results of a research program conducted by the Illinois Institute of Technology (IIT, Chicago) to determine the suitability and effectiveness of the lamella electrosettler -- a novel solid-liquid separation device -- for removing fine shale particles from shale oil via the application of an electric field. The work was conducted by IIT from November 1989 through December 1990 as a subcontractor to the Institute of Gas Technology (IGT, Chicago), which served as the prime contractor for a multifaceted research program sponsored by the U.S. Department of Energy (DOE) Morgantown Energy Technology Center/Laramie Project Office (METC/LPO) under Contract No. DE-AC21-87MC11089. The overall objective of the larger program was to develop the "Pressurized Fluidized-Bed Hydroretorting (PFH) Process for Eastern Oil Shales." The subtask undertaken by IIT was part of a larger task entitled "Testing of Process Improvement Concepts."

The lamella electrosettler has been shown to be an effective method for separating fine particulate (including colloidal) matter from a liquid using the application of an electric field. Using the walls of the settler as electrodes and during continuous operation, solids migrate preferentially toward one of the electrodes and become concentrated in the refuse stream. The product stream is clarified of particulates.

The success of the process depends upon the physical properties of the solids and liquids being tested. For example, a model system composed of kerosene and alumina particles (1 weight percent in the solution) was readily separated in the lamella electrosettler during tests conducted by IIT. The specific conductance of pure kerosene was determined by IIT to have a conductivity of about 8 X 10^{-11} mho/m. The specific conductance of samples of shale oil produced from Alabama and Ohio shale by IGT during testing of the PFH process were determined to be about 8 X 10^{-8} and 3.7 X 10^{-6} mho/m, respectively. The presence of polar compounds in the oil is thought to be responsible for the high conductivity.

A sample with a high specific conductance is not suitable for separation in the lamella electrosettler. The liquid begins to heat up under the influence of the electric field and, eventually, may short. Also, under these conditions, the particles cannot maintain a charge.

The high conductivity of the shale oil samples tested rendered them unsuitable for further testing in the lamella electrosettler.

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INTRODUCTION

The Devonian oil shales of the Eastern United States are a significant domestic energy resource. The overall objective of the 3-year program, initiated in October 1987 by the U.S. Department of Energy (DOE) under Contract No. DE-AC21-87MC11089, is to perform the research necessary to develop the pressurized fluidized-bed hydroretorting (PFH) process for producing oil from Eastern oil shales. The program also incorporates research on technologies in areas such as raw shale preparation, beneficiation, product separation, and waste disposal that have the potential of improving the economics and/or environmental acceptability of recovering oil from oil shales using the PFH process.

In order to accomplish all of the program objectives, the Institute of Gas Technology (IGT), the prime contractor, worked with seven other institutions: the University of Alabama/Mineral Resources Institute, Illinois Institute of Technology (IIT), the University of Michigan, the University of Nevada, Ohio State University, Tennessee Technological University, and the University of Pittsburgh.

This Topical Report on "Shale Oil Dedusting" presents the results of work conducted by IIT to determine the suitability and effectiveness of the lamella electrosettler -- a novel solid-liquid separation device -- for removing fine shale particles from shale oil via the application of an electric field. The work was conducted by IIT from November 1989 through December 1990. The subtask undertaken by IIT was part of a larger task entitled "Testing of Process Improvement Concepts."

TECHNICAL DISCUSSION

Background and Program Approach

Fluidized-bed processing of fine-size beneficiated shale has the potential to contaminate shale oil with fine-size shale particles. Thus, effective and efficient removal of fines from shale oil is very important. A novel approach that has shown promising results with coal was evaluated for the removal of fine particulates from shale oil.

All coal liquefaction processes require separation of fine particles entrained in the liquid product stream. The mineral matter content may range from 3 to 21 weight percent and particle sizes may be as small as 5 μm or less. To be considered effective, the solid separation processes must have good mechanical reliability and operability, and should add minimal energy requirements to the process. A number of separation processes have been used in the past, such as filtration, anti-solvent precipitation and sedimentation, critical solvent deashing, hydroclones, etc. The presence of fine colloidal particles makes these mechanical processes ineffective because they clog and block the path of clear liquid.

Two principal types of filtration are generally used: rotary drum precoat and pressure leaf. However, removal of fine colloidal particles is extremely difficult with filtration. Experimental work has been performed at the SRC (solvent refined coal) pilot plants in Wilsonville, Alabama, and in Ft. Lewis, Washington. The results of this work have eliminated the consideration of rotary drum and horizontal leaf pressure filters because these systems are batch-mode systems and have low mechanical reliability, which lead to poor performance and prohibitive economics for scale-up.[1]

The addition of anti-solvents to coal-derived liquids causes the asphaltene and pre-asphaltene fractions in the oil to precipitate. The precipitated asphaltene then agglomerates mineral matter causing the sedimentation rate to increase. This process was tested at the Ft. Lewis SRC pilot plant with erratic results.[2]

Hydroclones have been used in many processes for separating solids from liquids. The basic advantages of hydroclones are simplicity, economy, minimum maintenance, and continuous operation. The disadvantage lies in the fact that micrometer-size particles or colloidal particles cannot be removed effectively.

The vertical sedimentation process is often too slow, especially when the particles are small. Therefore, simple devices that could accomplish the solid-liquid separation more rapidly are needed. One such class of devices is known commercially as Lamella Settlers. An improved version of this device is a Lamella Electrosettler, which has been used by the Illinois Institute of Technology (IIT) to effectively separate coal fines from coal liquids. The concept may be applicable for removal of fines from shale oil. The development of an efficient method to remove the shale fines from shale oil will improve the overall quality of the product oil and the thermal efficiency of the process.

Lamella settlers are commonly used industrially for clarification of aqueous and nonaqueous slurries.[3] Acrivos et al.[4] presents a theory of their operation from a hyrodynamic point of view. Lamella settlers, however, cannot be used to remove particles from colloidal suspensions. Such suspensions form in nonaqueous liquids containing asphaltenes. These asphaltenes adsorb on the fine particles present in various liquids of interest in the production of synthetic fuels from coal, oil shale, and tar sands, as well as in some oil refining operations. Because the asphaltenes carry a surface charge, a nearly stable colloidal suspension is formed in the presence of a sufficiently high concentration of asphaltenes. Although several approaches can be used to separate these particles, it had been previously shown[5,6] that an effective means to remove such particles is to apply a high-voltage electric field. Previously a cross-flow electro-filter had been used. However, such a device is too expensive to be used in the production of reasonably priced fuels and does not produce a sufficiently concentrated slurry.

A lamella settler, with two of its walls being used as electrodes, is a much less expensive device and can, when operated continuously, produce a clarified liquid and a concentrated slurry. The operation and the hydrodynamic theory of a batch lamella settler with an applied electric field, which is called a "lamella electrosettler" is described. This device was tested at IIT with a tetralin slurry containing fine alumina particles stabilized with a commercial surfactant, Aerosol OT (dioctyl sodium sulfosuccinate). Such a system is a model for removal of particles from coal liquids or tar sand extracts, where the surfactant Aerosol OT replaces asphaltenes. This similarity was shown in sedimentation tests in cylindrical columns with and without an applied electric field in liquids containing high concentrations of asphaltenes.[7,8]

Subtask 3.4. Electroseparation of Fines From Shale Oil

The objective of this subtask was to determine the feasibility of using a lamella electrosettler for separating fine shale particles from shale oil. This task focused on the application of an electric field to separate the fines, which carry a significant surface charge due to adsorption of asphaltenes present in the oil. This subtask was further subdivided into the following subtasks: Charge Determination, Lamella Electrosettler Tests, and Design of a Continuous-Unit Hydrodynamic Model.

Subtask 3.4.1. Charge Determination

The objective of this subtask was to determine the surface charge of fines in various shale oil samples and to correlate this charge to other properties of the solid-liquid system, such as adsorption of shale asphaltenes and maltenes on the fines, the surface and interfacial tensions of the oils, and the effect of the presence of water.

A colloidal suspension consisting of a slurry of 1 vol % α-alumina particles dispersed in kerosene using 25 mmol/L Aerosol OT surfactant was identified as a model system. This model system was used for preliminary testing of a laboratory-scale continuous lamella electrosettler.

Pure kerosene has a very low specific conductance of about 8×10^{-11} mho/m. In previous work with alumina-tetralin systems, aerosol OT was used as a dispersant in settling experiments. The alumina particles were obtained from Buehler Corporation, with a narrow size distribution of 0.3 to 5 μm with an average particle size of 3 μm.

Specific conductance and electrophoretic mobility data were obtained using a Zetameter. Figure 3-1 shows the specific conductance of kerosene as a function of electric field strength at various aerosol OT concentrations. Although the specific conductance of kerosene was found to increase with an increase in electric field strength and aerosol OT concentrations, the actual value of specific conductance was very low. Therefore, kerosene was deemed to be suitable for use as a nonaqueous media for solid-liquid separation using a lamella electrosettler.

The specific conductance of a kerosene sample used for modeling tests was found to be about 10^{-10} mho/m. If the conductance of the sample was less than 10^{-10} mho/m, then the batch and continuous settling experiments could be conducted in the electrosettler. If the conductance was greater than this value, the sample might not be suitable for electrosettler tests.

The specific conductances of shale oils from Alabama (AL) (Test C-37) and Ohio (OH) (Test C-30) were measured to be 8.13 X 10^{-8} and 3.67 X 10^{-6} mho/m, respectively. The specific conductance of AL and OH shale oils were about 2 and 4 orders of magnitude higher, respectively, than that of the kerosene-aerosol sample used in previous settling experiments. The higher conductance of the shale oil samples is not favorable for the electroseparation of shale fines from the oil. For AL shale oil, the specific conductance is not a function of electric field strength. It had been previously determined (Figure 3-1) that the specific conductance of the kerosene was about 10^{-10} mho/m at low electric field strength and varied with the electric field strength.

Aging the sample of OH shale oil for 45 days increased the specific conductance from 3.67 X 10^{-6} to 4.49 X 10^{-6} mho/m. The increase in the specific conductance suggests that the shale oil was being oxidized and that the quantity of polar components in the sample was increasing.

Subtask 3.4.2. Lamella Electrosettler Tests

The objectives of this subtask were to study the settling rates and concentration of shale oil fines in various shale oils, using a lamella electrosettler. In particular, the effect of solids concentration and the configuration of the lamella electrosettler were to be studied. Laboratory-scale batch tests to study the settling characteristics of shale oil fines were to be conducted.

Experimental Equipment

A conventional lamella settler consists of inclined parallel plates stacked to form channels into which a slurry is fed for separation. The retention times of the slurries can be reduced by an order of magnitude or more below those in corresponding vertical settlers. One can interpret the increase in settling rates as due to an increase in the surface area available. Still, the lamella settler is not efficient in the removal of particles of colloidal size, which describes most of the shale oil fines.

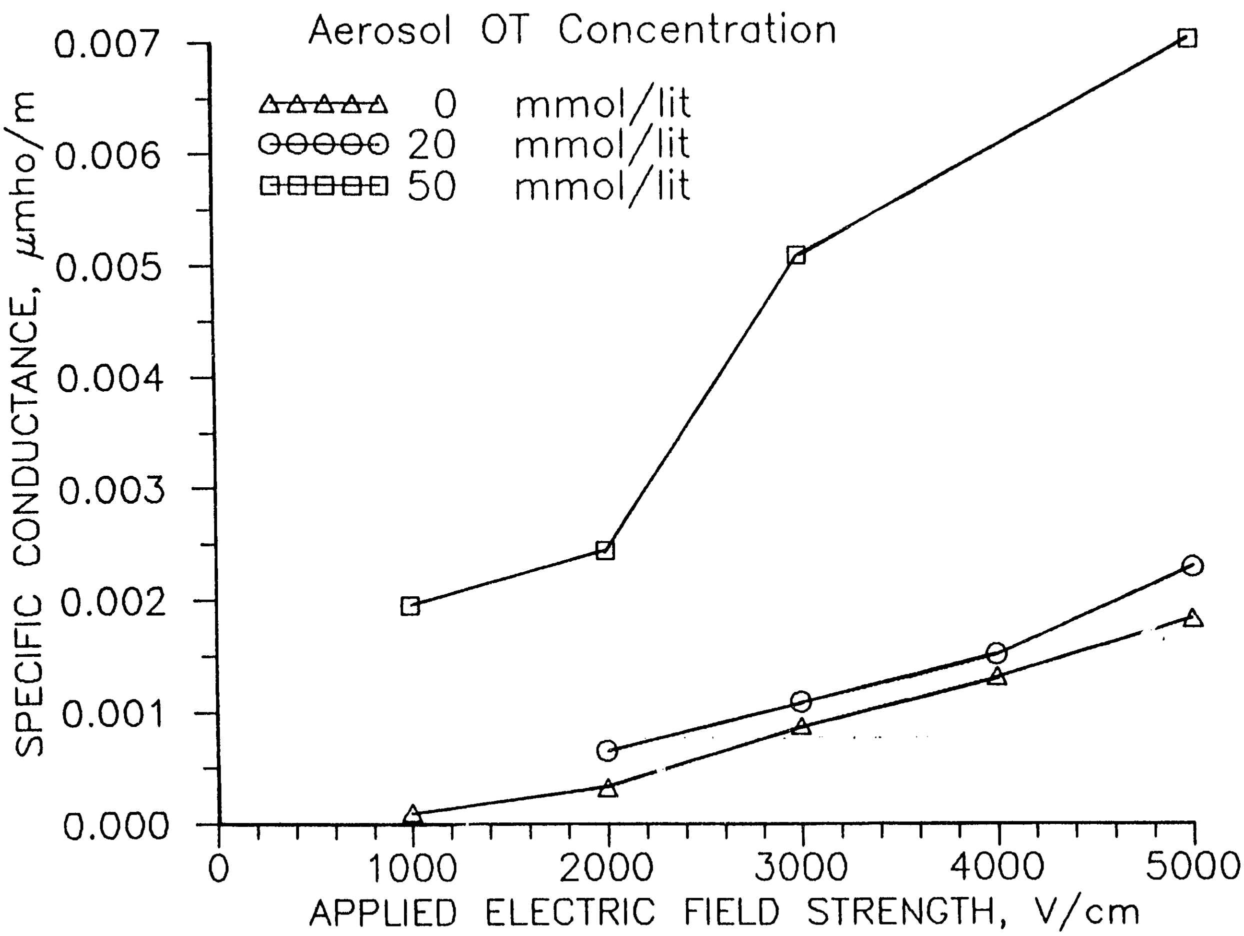

Figure 3-1. SPECIFIC CONDUCTANCE OF KEROSENE vs. ELECTRIC FIELD STRENGTH AT VARIOUS AEROSOL OT CONCENTRATIONS

However, if an external electric field is applied in the proper direction, efficient and effective removal of colloidal particles is possible. Figure 3-2 shows a schematic diagram of a lamella electrosettler. Two of the settler walls are used as electrodes to provide an external force on the charged particles. A high d-c voltage within the range of 1000 to 10,000 volts was applied across the electrodes. The settling rates and concentration profiles were studied for various angles of the lamella electrosettler. Since shale oils are opaque, the settling rates and concentration profiles were measured using an X-ray densitometer and an NaI scintillation detector.

The electrosettler constructed is 60 cm long, 6 cm wide, and 8.5 cm deep. Multiple feed inlets on the upper plate enable testing of various feed locations. Clear liquid overflows at the top of the inclined plate and sludge is removed at the underflow.

The electrodes (two 1-mm thick aluminum plates) are attached to the inside of the inclined walls of the settler. Up to 30 kV of d.c. voltage can be applied across the electrodes with the power supply. The settler is mounted on an adjustable frame to vary the angle of inclination during the tests. The slurry feed tank can be continuously agitated and pumped at a constant flow rate using a peristaltic pump.

The electrosettler is shown schematically in Figure 3-3. Testing began using a model system, 1 vol % alumina particles dispersed in kerosene with 25 mmol/L of surfactant. The objective of the tests was to determine the effects of applied voltage (250 to 1000 V/cm) and angle (40° to 60° measured from the vertical) on the settling characteristics of the model system. Upon application of the electric field, a sharp interface between the suspension and the particle-free liquid formed immediately. A visible boundary developed and the steady-state thickness was reached quickly. The height of the interface was recorded during the tests.

The results (Figure 3-4) show that increasing the applied voltage or decreasing the angle of inclination increased the sedimentation rate. For example, with an applied electric field of 250 V/cm and an angle of inclination of 50°, the settling time was about 64 minutes. Increasing the electric field to 1000 V/cm at the same angle, reduced the settling time to 9 minutes. Electrophoretic mobility data obtained in the batch tests agree with literature data.

Tests were also conducted with the lamella electrosettler in continuous mode with a product flow rate of 50 ml/min. At this flow rate, the steady interface heights (of the alumina particles) were lower at a 60° angle of inclination than at 40° across the range of applied voltages tested (see Figure 3-5). The height of the steady interface (H_s) can be estimated according to the equation --

$$H_s = [F_{\cup f}/U_e w \cdot \cos(\alpha) - b\sin(\alpha)] \qquad (3\text{-}1)$$

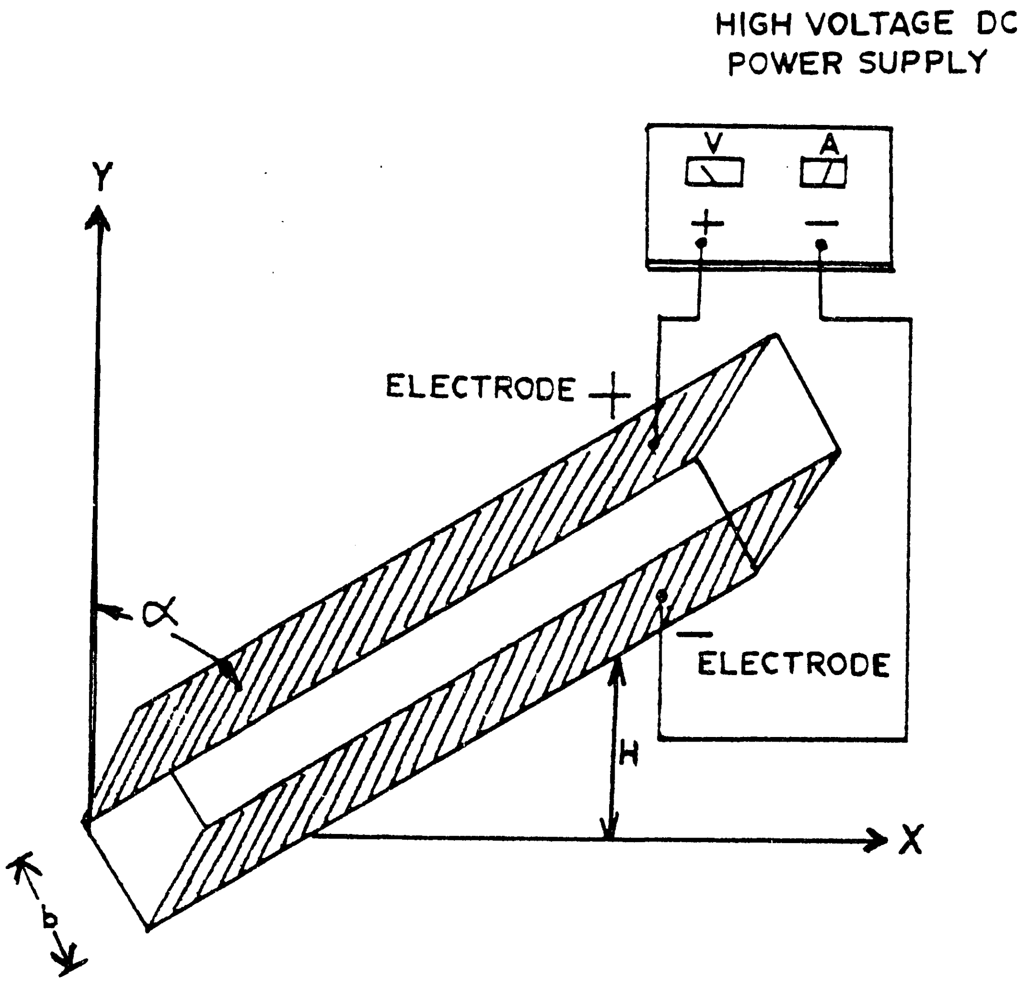

Figure 3-2. LAMELLA ELECTROSETTLER

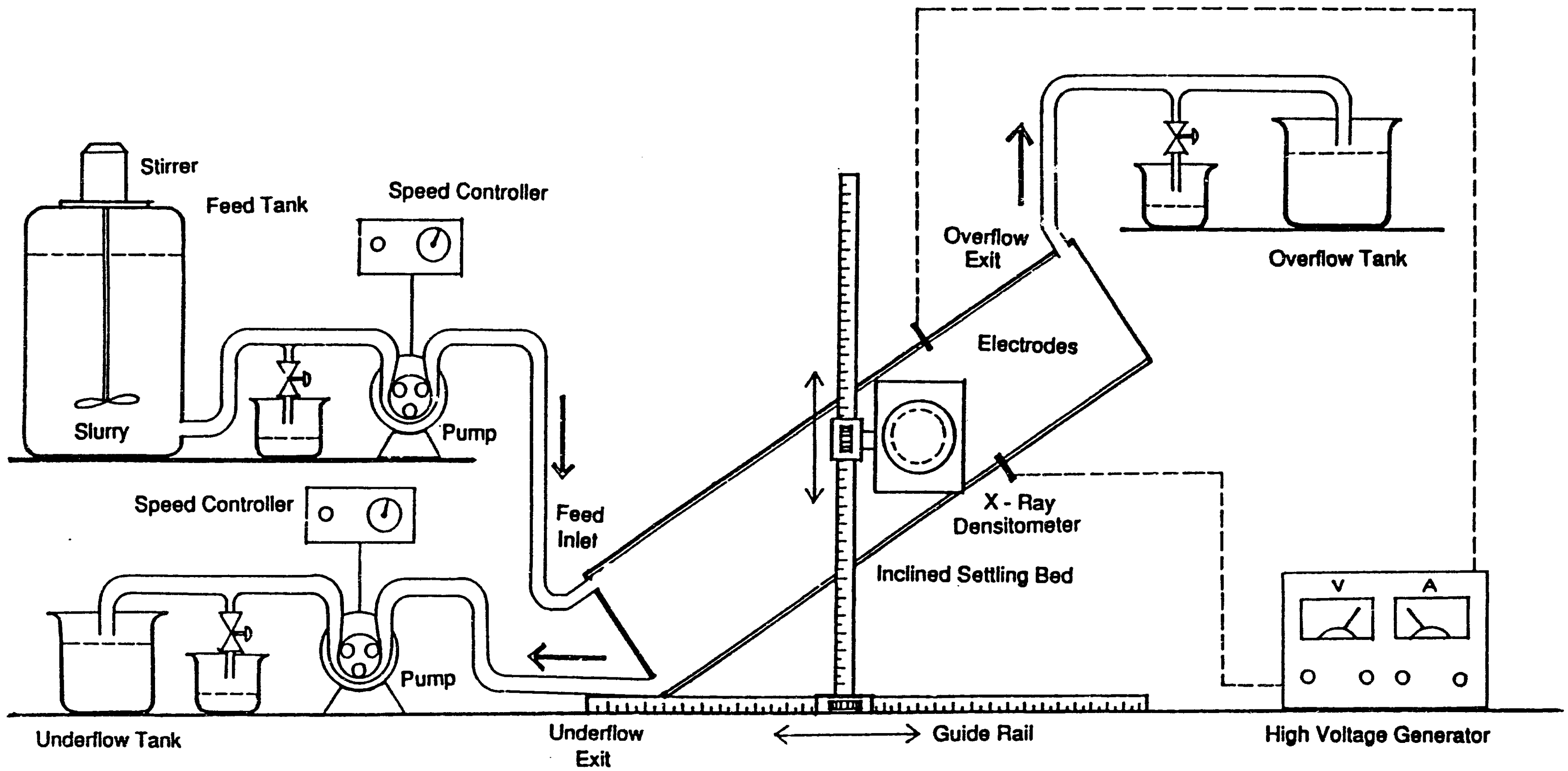

Figure 3-3. CONTINUOUS LAMELLA ELECTROSETTLER SETUP

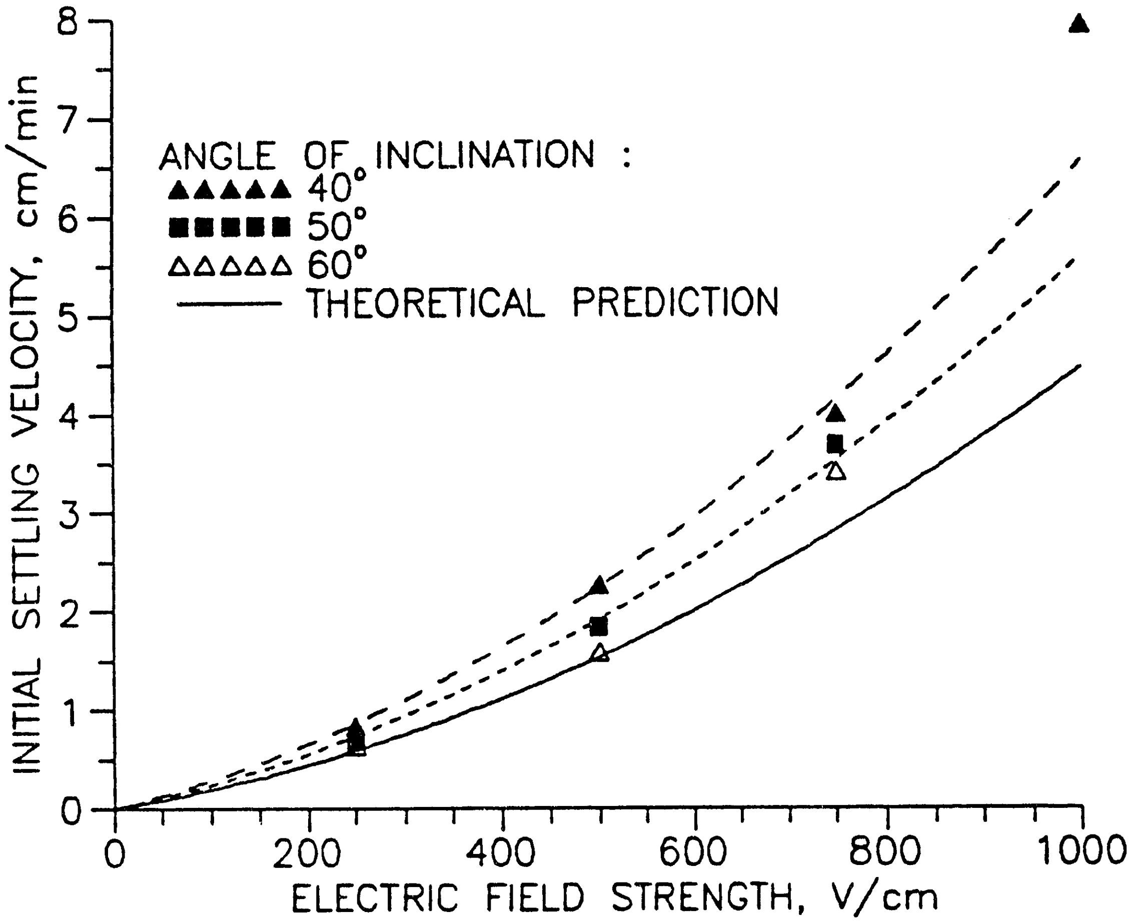

Figure 3-4. EFFECT OF ELECTRIC FIELD STRENGTH AND ANGLE OF INCLINATION ON INITIAL SETTLING VELOCITY IN A LAMELLA ELECTROSETTLER

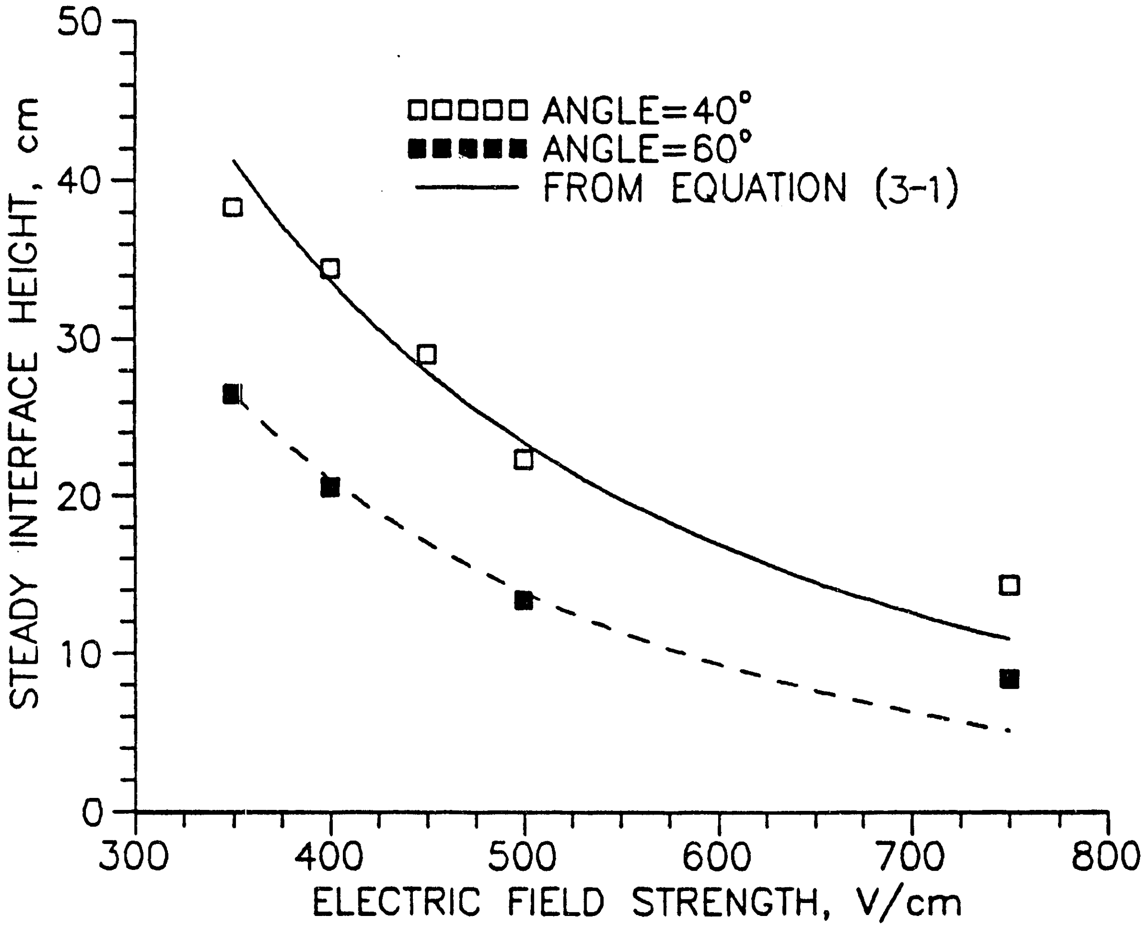

Figure 3-5. STEADY INTERFACE HEIGHT vs. ELECTRIC FIELD STRENGTH IN THE CONTINUOUS LAMELLA ELECTROSETTLER FOR AN OVERFLOW RATE OF 50 ml/min

where --

U_e is the electrophoretic velocity

F_{of} is the volumetric flow of clarified liquid in the overflow

w is the depth of the electrosettler in the -z direction

α is the angle of inclination

The equation was fitted to the data with a value of "w" equal to 4, rather than the actual bed depth of 8.5 cm. The reason for the difference is due primarily to the recirculation of fine particles in the suspension region. Hence, the applicability of the formula is limited to low feed flow rates.

Subtask 3.4.3. Design of a Continuous-Unit Hydrodynamic Model

The use of a hydrodynamic model permits scale-up of a continuous process. The objective of this subtask was to design a continuous solid-liquid separating unit using the hydrodynamic model to optimize design and operating conditions for various shale oils.

Clean kerosene was separated from a model system of alumina/kerosene. The specific conductance for the model system was 10^{-10} mho/m.

The lamella electrosettler was not suitable for the separation of fines from an oil with a specific conductance greater than 10^{-10} mho/m. A large quantity of polar compounds in the raw shale oils was postulated as the reason for the high specific conductance of the samples. A sample with a high specific conductance causes the samples to heat up and does not allow the shale fines to maintain a charge. No further tests were conducted using shale oil due to the high specific conductance of AL and OH shale oils.

ACKNOWLEDGMENT

IGT acknowledges the significant work conducted on this project by IIT Professors D. T. Wasan and D. Gidaspow, their principal investigators, and colleagues.

REFERENCES CITED

1. Deutsch, D. J., "Solid Liquid Separation Route Are Nearing Demonstration Stage," Chem. Eng., 33, (1980) August 25.

2. Henry, J. D., Jr., Prudich, M. E. and Vaidynathan, K. R., "Novel Separation Process for Solid/Liquid Separation in Coal Derived Liquids," Sup. and Purification Methods, 8(2), 81-118, 1979.

3. Forsell, B. and Hedstorm, B., "Lamella Sedimentation: A Compact Separation Technique," Journal WPCF 47, 4, 834-42, 1975.

4. Acrivos, A. and Herbolzheimer, E., "Enhanced Sedimentation in Settling Tanks With Inclined Wall," J. Fluid Mech., 92, 435-57, 1979.

5. Lee, C., Gidaspow, D. and Wasan, D. T., Proceedings of the Technical Program of the International Powder and Solids Handling and Processing, Philadelphia, 528-34, May 1979.

6. Lo, Y. S., Gidaspow, D. and Wasan, D. T., "Separation of Colloidal Particles From Nonaqueous Media by Cross-Flow Filtration," Separation Sci. & Tech. 18, 1323-49, 1983.

7. Shih, Y. T., Gidaspow, D. and Wasan, D. T., "Sedimentation of Fine Particles in Nonaqueous Media" Colloids and Surfaces 21, 393-429, 1987.

8. Shih, Y. T., "Hydrodynamics of Separation of Particles: Sedimentation and Fluidization," Ph.D. Thesis, Illinois Institute of Technology, Chicago, 1986.

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