DOE/PC/91164--T1

TECHNICAL PROGRESS REPORT

DOE/PC/91164--T1

DE93 007671

for the Fourth Quarter

(July 1, 1992 - September 30, 1992)

DEVELOPMENT OF THE SELECTIVE HYDROPHOBIC COAGULATION PROCESS

by

R.-H. Yoon and G. H. Luttrell

Virginia Center for Coal and Minerals Processing
Virginia Polytechnic Institute and State University
Blacksburg, Virginia 24061-0258

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.

Contract Number:

DE-AC22-91PC91164

Project Manager:

Richard Read
U.S. Department of Energy
Pittsburgh Energy Technology Center
P. O. Box 10940
Pittsburgh, Pennsylvania 15236

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

ABSTRACT

A novel technique for selectively coagulating and separating coal from dispersed mineral matter has been developed at Virginia Tech. The process, *Selective Hydrophobic Coagulation (SHC)*, has been studied since 1986 under the sponsorship of the U.S. Department of Energy (Contracts DE-AC22-86PC91221 and DE-AC22-90PC90174). The *SHC* process differs from oil agglomeration, shear or polymer flocculation, and electrolytic coagulation processes in that it does not require reagents or additives to induce the formation of coagula. In most cases, simple pH control is all that is required to (i) induce the coagulation of coal particles and (ii) effectively disperse particles of mineral matter. If the coal is oxidized, a small dosage of reagents can be used to enhance the process.

The technical work program was initiated on July 1, 1992, following DOE/NEPA approval of the project in late May. For Task 2, force-distance curves were generated for DDOA Br coated mica surfaces in water and used to calculate hydrophobicity constants and decay lengths for this system (Task 2.1.1); and a new device for the measurement of water contact angles, similar to the Wilhelmy plate balance, has been built for use in Task 2.1.2. For Task 3, 225 kg samples of Pittsburgh No. 8 and Elkhorn No. 3 seam coals were obtained (Task 3.1); a static mixer test facility for the study of coagula growth was set up and was undergoing shakedown tests at the end of the quarter (Task 3.2.1); a bench-scale lamella thickener was being constructed (Task 3.3.1); and preliminary coagula/mineral separation tests were being conducted in a bench-scale continuous drum filter (Task 3.3.3).

TABLE OF CONTENTS

ABSTRACT	i
TABLE OF CONTENTS	ii
LIST OF FIGURES	iii
LIST OF TABLES	iii
PROJECT OBJECTIVES	1
PROJECT TASKS	1
Task 1 - Project Planning	1
Task 2 - Fundamentals	1
2.1 <u>Parameter Estimation for the Hydrophobic Interaction Energy</u>	
<u>Function</u>	1
2.1.1 <i>Surface Force Measurements</i>	3
2.1.2 <i>Hydrophobicity Measurements</i>	8
2.1.3 <i>Parameter Estimation</i>	10
Task 3 - Process Development	13
3.1 <u>Sample Acquisition</u>	13
3.2 <u>Mixing Studies</u>	15
3.2.1 <i>Agitation Using In-Line Mixers</i>	15
3.3 <u>Advanced Separation Methods</u>	16
3.3.1 <i>Lamella Thickener</i>	18
3.3.3 <i>Vacuum Filtration</i>	18
REFERENCES	21

LIST OF FIGURES

Figure 1:	Force-distance curves measured between two DDOA Br-coated mica surfaces in nanopure water at distances of 10-40 nm. The straight lines are the best-fit exponential curves.	5
Figure 2:	Force derivative-distance curves measured between two DDOA Br-coated mica surfaces in nanopure water at distances of 30-120 nm. The straight lines are the best-fit exponential curves.	6
Figure 3:	Apparatus for the measurement of water contact angles on mica sheets coated with carbon. Sample(1), balance arm (2), vessel containing water (3), chamber containing water(4).	9
Figure 4:	The dependence of the force parameter C_1 (Eq. [2]) on contact angle. The curve is Eq. [15].	14
Figure 5:	Schematic representation of experimental set-up for static mixer studies. . .	17
Figure 6:	Schematic drawings of the bench-scale lamella thickener for the separation of coal coagula from dispersed mineral particles.	19

LIST OF TABLES

Table I:	Experimental values of the parameters of the short- and long-range attraction (Eqs.[3] and [5]) of mica covered by a monolayer of DDOA.	7
----------	---	---

PROJECT OBJECTIVES

The two earlier projects established the fundamental mechanisms of the *SHC* process, identified the various parameters that affect its operation, and investigated several methods, i.e., sedimentation, elutriation and drum filtration, for separating the coal coagula from dispersed mineral matter. The main objectives of the current project are to 1) determine the parameters necessary for developing an extended DLVO theory that can be used for predicting the optimum conditions of the *SHC* process, and 2) continue the development of methods for improving the coagulation process (by increasing the efficiency of mixing) and for separating coal coagula from dispersed mineral matter.

PROJECT TASKS

Task 1 - Project Planning

The technical work program was initiated on July 1, 1992, following DOE/NEPA clearance in late May, and preliminary approval of the draft work plan. The final project work plan was approved by the DOE's Contracting Officer's Representative on August 14, 1992.

Task 2 - Fundamentals

2.1 Parameter Estimation for the Hydrophobic Interaction Energy Function

The major driving force for the *SHC* process is the hydrophobic interaction energy (V_H). In general, this term decreases exponentially with the distance (H) separating two interacting particles according to the relationship:

$$V_H = \frac{a_1 a_2}{(a_1 + a_2)} C D_o \exp\left(\frac{H}{D_o}\right) \quad [1]$$

in which a_1 and a_2 are the particle diameters, and C and D_o are the hydrophobicity constant and decay length, respectively. In order to predict V_H , it is necessary to develop a method of estimating the values of C and D_o .

In the present work, the C and D_o parameters are being determined with an Anutech Mark IV Surface Force Apparatus. Unfortunately, coal samples cannot be used for the direct force measurements because it is not possible at this point to prepare a molecularly smooth coal surface. For this reason, the surface force measurements are being conducted using silica and mica plates. The hydrophobicity of silica is being controlled by reacting the surface with either trimethylchlorosilane (in solution or gas phase) or octadecyltrichlorosilane in cyclohexane. The extent of methylation is being monitored using Fourier Transform Infrared spectroscopy (FTIR) and water contact angles. The optimum reaction conditions will be determined from these measurements. Mica surfaces are being made hydrophobic by:

- 1) adsorption of dodecylamine from aqueous solution,
- 2) deposition of dimethyldioctadecylammonium (DDOA) bromide using a Langmuir trough,
- 3) self-adsorption of octadecyltrimethoxysilane from tetrahydrofuran or other suitable organic solvent, or
- 4) vacuum deposition of carbon.

The method employed depends on the degree of hydrophobicity required. The criteria used to choose the surfaces will include:

- 1) stability of the hydrophobizing layer in water and salt solutions,
- 2) measurements of water contact angle, and/or
- 3) number density of hydrocarbon chains as measured by FTIR.

As with the silica surfaces, the extent of hydrophobization of the mica surfaces is being measured by FTIR and water contact angles. The hydrophobicity of each of these surfaces is also being characterized by means of the non-dispersion component (W_a^{nd}) of the work of adhesion of water. These values are being determined using the well-known octane-in-water contact angle technique.

Once the values of C and D_o have been determined for a range of W_a^{nd} values, regression analyses will be conducted to derive the functional relationships between:

- 1) C and W_a^{nd} , and
- 2) D_o and W_a^{nd} .

These relationships will then be used for estimating the values of C and D_o from the value of W_a^{nd} for coal. The procedure of determining these relationships is different from that employed in the previous project (DE-AC22-90PC90174) in that it relies on direct surface force measurements.

2.1.1 Surface Force Measurements

Using the Anutech Mark IV Surface Force Apparatus, the hydrophobic attraction has been investigated as a function of the shortest distance (H) between two mica cylinders of radius R , hydrophobized by dimethyldioctadecylammonium (DDOA) bromide. The Langmuir-Blodgett method was used for the deposition of surfactant. The deposition was made at two different surface pressures (Π_s) = 25 and 35 mN/m. The surface area per head

group was equal to 0.50 and 0.44 nm², respectively.

The force (F) between the cylinders and its derivative (dF/dH) are shown in Figs. 1 and 2. The straight lines are the best exponential fits.

According to Derjaguin (1934), the force (F) between cylinders is related to the specific (per unit area) surface energy (E) of the flat surface as follows:

$$F/R=2\pi E \quad [2]$$

Fig. 2 implies that this force can be best described by an equation with two exponential terms, i.e.:

$$-F/R=C_1\exp(-H/D_1)+C_2\exp(-H/D_2) \quad [3]$$

where the coefficients C_1 and C_2 characterize the force strengths and D_1 and D_2 characterize the decay lengths, i.e. the rate of force decay with distance. Usually, for mica covered by hydrocarbon surfactants, $D_1 \approx 1-2$ nm and $D_2 \approx 10-25$ nm. These will be described as "short range" and "long range" forces, respectively, although both terms are "long range" when compared with the intermolecular distance.

Combining Eqs.[2] and [3] gives the formula for the specific surface energy (E):

$$E=-\frac{C_1}{2\pi}\exp(-H/D_1)-\frac{C_2}{2\pi}\exp(-H/D_2) \quad [4]$$

All four parameter values can be obtained from Fig. 2 using the differential form of Eq.[3]. i.e.:

Figure 1: Force-distance curves measured between two DDOA Br-coated mica surfaces in nanopure water at distances of 10-40 nm. The straight lines are the best-fit exponential curves.

Figure 2: Force derivative-distance curves measured between two DDOA Br-coated mica surfaces in nanopure water at distances of 30-120 nm. The straight lines are the best-fit exponential curves.

$$dF/RdH=(C_1/D_1)\exp(-H/D_1)+(C_2/D_2)\exp(-H/D_2) \quad [5]$$

The Derjaguin approximation (Eq. [2]) yields the relationship between dF/dH and the disjoining pressure (Π):

$$\Pi = -\frac{dE}{dH} = -\frac{dF}{2\pi R dH} \quad [6]$$

Combining Eqs.[5] and [6] gives:

$$\Pi = -\frac{C_1}{2\pi D_1}\exp(-H_1/D_1) - \frac{C_2}{2\pi D_2}\exp(-H/D_2) \quad [7]$$

These equations are used in Section 2.1.3 below.

Only two parameter values (C_2 and D_2) could be obtained from Fig. 1, because it was not possible to measure the force at distances less than 10 - 15 nm, where the first term of Eq.[3] acts.

Table I summarizes the parameter values obtained from this analysis and compares

Table I: Experimental values of the parameters of the short- and long-range attraction (Eqs.[3] and [5]) of mica covered by a monolayer of DDOA.

Method	C_1 (mN/m)		D_1 (nm)		C_2 (mN/m)		D_2 (nm)	
	I	II	I	II	I	II	I	II
dF/dH	331	520	2.07	1.80	1.25	1.65	25	13.2
F	-	-	-	-	-	4.0	-	30

Column I gives results from Tsao *et al.* (1991), column II gives the current results.

these with results presented by Tsao, *et al.*, (1991).

In principle, the force parameter values should be independent of the measurement method used (force or pressure). However, it is obvious from Table 1 that there are differences in the parameter values obtained by the two different methods, probably because the forces and force derivatives (pressure) were measured at different distances. This may also mean that the dependence of force and of pressure on distance includes not two, but three or more exponential terms.

2.1.2 Hydrophobicity Measurements

A new apparatus, similar to the Wilhelmy plate balance, was constructed for the measurement of contact angles. This device is shown in Fig. 3. The sample (1), a freshly cleaved, 10x20mm rectangular sheet of mica coated with carbon, hangs from an automatic balance arm (2) and dips into vessel (3), which is filled with water. Vessel (3) is connected to chamber (4) with flexible tubing. Changing the vertical position of this chamber causes water to flow in and out of vessel (3), providing an advancing/receding water meniscus on the mica sample. The meniscus velocity is about 50 $\mu\text{m/s}$ - in this case the angles obtained are independent of the velocity. The apparatus is installed in an air swept, temperature controlled enclosure that provides a temperature range of 25 - 50°C.

The contact angles are calculated from the formula:

$$\cos\theta_{a,r} = F/(2\gamma d) \quad [8]$$

where indices *a* and *r* relate to the advancing and receding angles, *d* is the sample width (usually about 10 mm), *F* is the measured force (caused by the liquid meniscus acting on the

Figure 3: Apparatus for the measurement of water contact angles on mica sheets coated with carbon. Sample(1), balance arm (2), vessel containing water (3), chamber containing water(4).

sample), and γ is the surface tension of water (72 mN/m). Eqn. [8] neglects the Archimedean force because of the small sample thickness.

This method has advantages over the more common droplet method because it provides better measurement precision for small changes in contact angle, and because the results are averaged over the width of the sample. Additionally, the method can be used to yield results vertically across the surface of the sample. The apparatus is especially convenient for the measurement of contact angles on thin flat samples.

The results of the contact angle measurements and the relationships between the contact angle and the force parameters will be presented in the next report.

2.1.3 Parameter Estimation

The theoretical relationship between the force parameter, C_l , in Eq. [3], i.e. the nondispersion component of the work of adhesion (E_a^{nd}), and the contact angle is derived below. If the contact angle (θ) is used as the measure of the degree of hydrophobicity of the sample, then the hydrophobic force can be related to θ through the well-known Young's equation:

$$\gamma_{sl} = \gamma_{sv} - \gamma_{lv} \cos \theta \quad [9]$$

where γ is the interfacial tension and indices s , l and v refer to the solid, liquid and vapor, respectively. The value of γ_{sv} can be obtained from measurements of the adhesion force between solids in air (see below). The value of γ_{sl} is related to the specific free energy (E_{sl}) of the flat solid at the solid/liquid interface (i.e., when the work of adhesion (E_a) = $-E_{sl}$), as follows:

$$\gamma_{sl} = -E_{sl}(H_0)/2 \quad [10]$$

where H_0 is intermolecular distance, i.e., $H_0 \approx 0.3$ nm. The distance H_0 is not zero due to Born repulsion, which compensates for the action of the surface forces.

The energy (E_{sl}) can be defined as the sum of three components: molecular/dispersive (E_d), ion-electrostatic (E_e), and hydrophobic (E_h):

$$E_{sl} = E_d + E_e + E_h \quad [11]$$

If it is assumed that the surface potential is constant when surfaces approach each other, then E_e (at $H=H_0$) is negligible when compared with the other components of the energy.

The molecular component of energy can be calculated as follows (Djaloshinsky, *et al.* 1961):

$$E_d = -\frac{A_{sls}}{12\pi H^2} \equiv -\frac{A_{131}}{12\pi H^2} \quad [12]$$

where A is the Hamaker constant for solid in liquid.

The energy E_h can now be calculated from Eq. [4], if the parameters C_1 , C_2 , D_1 and D_2 are known. If Eqs. [3] and [4] are valid down to intermolecular distances and assuming that $D_1 \gg H_0$ then, according to Eq.[4], only the value of C_1 is required for the calculation of $E_h(H_0)$, which is used in Eq. [10]:

$$E_h(H=0) = -C_l/2\pi \quad [13]$$

In this case, from [10] - [13] :

$$\gamma_{sl} = \frac{C_l}{4\pi} + \frac{A_{sls}}{24\pi H_0^2} \quad [14]$$

Substituting [14] in Young's Eq. [9] and solving this equation for C_l gives Eq. [15], which relates the value of the hydrophobic force parameter C_l to the contact angle θ :

$$C_l = 4\pi(\gamma_{sv} - \gamma_{lv}\cos\theta) - \frac{A_{sls}}{6H_0^2} \quad [15]$$

where, for water, $\gamma_{lv} = 72$ mN/m. According to Israelachvili and Adams (1978) and Rabinovich and Charaev (1990), the following values of A_{sls} can be used in Eq.[15]: $A_{sls} = 2.2 \cdot 10^{-20}$ J for mica in water and $A_{sls} = 1.1 \cdot 10^{-20}$ J for methylated silica in water (in the latter case it is assumed that the methyl layer is too thin to affect the Hamaker constant).

As discussed above, the value of γ_{sv} , can be obtained from adhesion force measurements on samples in air: $\gamma_{sv} = 27$ mN/m for mica hydrophobized by the hydrocarbons (Wu, 1982; Claesson *et al.*, 1986), and $\gamma_{sv} \geq 20$ mN/m (Yaminsky, *et al.*, 1975). The value of γ_{sv} can be also evaluated theoretically if it is assumed that it is affected only by the dispersion force, i.e., if:

$$\gamma_{sv} = \frac{A_{ss}}{24\pi H_0^2} \quad [16]$$

where A_{ss} is the Hamaker constant for two solids in vapor. Values for A_{ss} are given by Rabinovich, *et al.*, (1990): $A_{ss} = 1.1 \times 10^{-19}$ J for mica in air and $A_{ss} = 0.8 \times 10^{-19}$ J for silica. In this case the values of γ_{sv} calculated from Eq. [16] agree with the above mentioned values of γ_{sv} , obtained from the adhesion forces if the reasonable value of $H_0 = 0.23$ nm is used in Eq. [16].

A graph of C_1 vs. contact angle θ , plotted for the theoretical Eq. [15], is shown in Fig. 4. Theoretical relationships between other force parameters (D_1 , C_2 and D_2) and the contact angle cannot be derived. However, these relationships can be obtained from experimental data and from the literature. This will be done in subsequent reports.

Task 3 - Process Development

3.1 Sample Acquisition

The process development test work is being conducted on run-of-mine Pittsburgh No. 8 and Elkhorn No. 3 coal samples. The Pittsburgh No. 8 sample was obtained from a CONSOL Inc. property in northern West Virginia (CONSOL Sample Designation #LL-1022-45-1); the Elkhorn No. 3 sample from United Coal Company's Wellmore # 8 plant in Buchanan County, VA. Approximately 225 kgs (500 lbs) of each coal were collected and shipped to Virginia Tech in sealed 55-gallon drums. After delivery, each of the coals was pulverized to - 6.35 mm (1/4 inch) using a pilot-scale roll crusher, split into representative lots of approximately 1 kg each, transferred to air-tight plastic containers, flushed with

Figure 4: The dependence of the force parameter C_l (Eq. [2]) on contact angle. The curve is Eq. [15].

nitrogen, and stored in a freezer at -20°C to minimize oxidation.

3.2 Mixing Studies

One of the more important operating parameters to be examined in the present study is mixing intensity. A proper level of mixing is necessary in order to (i) increase particle-particle collision rates and (ii) provide sufficient kinetic energy to overcome the activation energy barrier (E) between the particles. Excessive mixing will result in an unnecessarily high energy consumption and a higher coagula breakage rate.

In the previous project (DE-AC22-90PC90174), a variety of mixing studies were carried out using a standard turbine impeller and baffled tank. In the follow-up work being conducted in this project, two other types of mixing devices are being examined, i.e., an in-line static mixer and an ultrasonic horn. These devices have the potential to offer a higher level of energy efficiency and improved control of mixing intensity. For each device, the intensity of mixing will be determined and used to estimate the kinetic energies of the particles in the suspension. The particle kinetic energies will then be compared with the activation energies (E) predicted from first principles in Task 2. This comparison will provide a means of verifying the optimization procedure to be conducted in Task 2.3.

3.2.1 Agitation Using In-Line Mixers

In this task, the use of in-line static mixers to provide the agitation required in the *SHC* process is being investigated. Micronized Elkhorn No. 3 seam coal slurries will be circulated through different in-line static mixers for a given period of time. The grind size, slurry percent solids and pH will be held constant at appropriate levels determined from

preliminary batch tests. When mixing is complete, the resultant coagula size distribution will be determined using a Lasentec particle size analyzer. The effects of mixer design, mixer geometry, slurry flow rate, and mixing time on the coagula size distribution will be investigated. Energy inputs will be calculated from the slurry flow rate and the pressure drop across the static mixer, according to the relationship:

$$E = QP \quad [17]$$

Where, E is the theoretical energy consumed, in watts (W)

Q is the flowrate, in m^3/sec

P is the pressure drop, in Pascals (Pa)

From the test data, a relationship between the mean coagulum size and the specific energy input will be derived. The kinetic energies of the particles will also be calculated and compared with the activation energies (E) determined in Task 2.2.

In-line static mixers have been purchased from the Koflo and Kenics Corporations and have been incorporated into the experimental set-up shown schematically in Fig. 5. The Lasentec particle-size analyzer probe is being used to measure coagula size distributions at different heights in the column from the bottom up to the point where the size distributions become stable. Shakedown testing of this unit was underway at the end of the quarter.

3.3 Advanced Separation Methods

One of the major advantages of the *SHC* process is that it does not require the use of agglomerating agents such as hydrocarbon oils or flocculants. However, the coagula formed by the *SHC* process are relatively weaker than those formed when agglomerating agents are

Figure 5: Schematic representation of experimental set-up for static mixer studies.

used. As a result, careful attention must be given to the proper selection and design of systems to recover these coagula. In the present work, four different methods of improving the recovery of coagula will be explored. These include the use of (i) a lamella thickener, (ii) a vacuum filter, (ii) a centrifuge, and (iv) a froth flotation cell. Each technique will be tested using bench-scale continuous units at throughputs of 0.25-0.5 kg/hr (0.5-1.0 lb/hr). These continuous separators will be tested at the optimum reagent conditions established from batch tests carried out on each of the two coals.

3.3.1 Lamella Thickener

A prototype lamella thickener is being constructed from Plexiglas for this task. Design information for this unit is given in Fig. 6. The unit includes provisions for adding countercurrent wash water to the lower section of the separator as a means of minimizing the entrainment of fine mineral matter. In principle, this device similar to the sedimentation/elutriation device employed during the previous project (DE-AC22-90PC90174), although the lamella design should offer significant improvements in throughput. The operating parameters to be examined in the present work will include feed percent solids, feed flow rate and wash water flow rate. Both coals will be tested in this device.

3.3.3 Vacuum Filtration

Vacuum filtration may have an advantage over the sedimentation techniques in that it is relatively unaffected by the slow sedimentation rate of the coal coagula. The applicability of vacuum filtration for the recovery of coagula will be studied by conducting tests using a bench-scale continuous filtration unit. Effects of filter medium, applied vacuum, feed percent solids, feed rate and wash water rate will be studied on both base coals. The

Plate Surface Area = $A = 60$ square inches

Number of plates = $n = 4$

Distance between plates = 175 inches

Cell Volume = 720 cubic inches

Plate Angle (α) for the Cell is adjustable from 30 to 55 degrees

→ plate angle when using wash water = 45 to 55 degrees

→ plate angle without wash water = 30 to 40 degrees

Effective Area = $n A \cos(\alpha)$

→ Effective Area using wash water = $4(60)\cos(50^\circ) = 154$ square inches

→ Effective Area w/o wash water = $4(60)\cos(35^\circ) = 197$ square inches

Figure 6: Schematic drawings of the bench-scale lamella thickener for the separation of coal coagula from dispersed mineral particles.

Center's continuous 100mm (4") x 200mm (8") diameter drum filter has been modified to permit the use of wash water to remove entrained mineral matter from the filter cake.

Preliminary pH control test work with the Elkhorn #3 coal sample is underway.

REFERENCES

- Israelachvili, J.N., and Adams, G.E., 1978. *J. Chem. Soc. Faraday Trans 1* **74**, 1975.
- Derjaguin, B.V., 1934. *Kolloid Z.* **69**, 155.
- Tsao, Y., Yang, S.X., Evans, D.F., and Wennerstrom, H., 1991. *Langmuir* **7**, 3154.
- Djaloshinsky, I.E., Lifshitz, E.M., and Pitaevsky, L.P., 1961. *Uspehi fizicheskikh nauk* (English translation) **73**, 381.
- Rabinovich, Ya.I., and Churaev N.V., 1990. *Colloid J. of USSR* **52**, 3029.
- Wu, S., 1982. "Polymer Interface and Adhesion", Dekker, New York.
- Claesson, P.M., Blom, C.E., Herder, P.C., and Ninham, B.W., 1986. *J. Colloid Interface Sci.* **114**, 234.
- Yaminsky, V.V., Yusupov, R.K., Amelina, E.A., Pchelin, V.A., and Shchukin, E.D., 1975. *Colloid J. of USSR* **37**, 918.

END

DATE
FILMED
3/19/93