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Development of a Gas-Promoted Oil Agglomeration Process

Final Report
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DEVELOPMENT OF A GAS-PROMOTED OIL AGGLOMERATION PROCESS

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ABSTRACT

The preliminary laboratory-scale development of a gas-promoted, oil agglomeration process for cleaning coal was carried out with scale model mixing systems in which aqueous suspensions of ultrafine coal particles were treated with a liquid hydrocarbon and a small amount of air. The resulting agglomerates were recovered by screening. During a batch agglomeration test the progress of agglomeration was monitored by observing changes in agitator torque in the case of concentrated suspensions or by observing changes in turbidity in the case of dilute suspensions. Dilute suspensions were employed for investigating the kinetics of agglomeration, whereas concentrated suspensions were used for determining parameters that characterize the process of agglomeration. A key parameter turned out to be the minimum time t_e required to produce compact spherical agglomerates. Other important parameters included the projected area mean particle diameter of the agglomerates recovered at the end of a test as well as the ash content and yield of agglomerates.

Batch agglomeration tests were conducted with geometrically similar mixing tanks which ranged in volume from 0.346 to 11.07 liters. Each tank was enclosed to control the amount of air present. A variable speed agitator fitted with a six blade turbine impeller was used for agitation. Tests were conducted with moderately hydrophobic Pittsburgh No. 8 coal and with more hydrophobic Upper Freeport coal using either n-heptane, i-octane, or hexadecane as an agglomerant.

It was shown that gas bubbles have to be present in an agitated system used for agglomerating a moderately hydrophobic coal. Gas bubbles trigger the process of agglomeration and participate in a very complex mechanism involving the interaction of particles, oil droplets, and gas bubbles. The process takes place in stages involving dispersion of oil and gas, flocculation, coagulation, and agglomerate building.

A study of the agglomeration kinetics of coal particles in dilute suspensions showed that the rate of agglomeration is proportional to the particle number concentration raised to a power between 1.0 and 1.3. The rate is also affected by particle hydrophobicity and the relative amounts of gas and agglomerant employed.

To determine the important characteristics of the process, numerous agglomeration tests were conducted with two kinds of coal in concentrated suspensions using i-octane as an agglomerant. A wide range of experimental conditions was explored in order to determine the effects of the following variables on the characteristic parameters: i-octane concentration, air concentration, particle concentration, tank diameter, impeller diameter, and impeller speed. For each kind of coal, the results were fitted by empirical equations to provide good to excellent correlations for the minimum time required to produce spherical agglomerates and for the final agglomerate diameter. In addition, the results provided a basis for size scale up of an agglomeration system.

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EXECUTIVE SUMMARY

The preliminary laboratory-scale development of a gas-promoted, oil agglomeration process for cleaning coal was carried out using scale model mixing systems representative of good industrial practice. In this process ultrafine coal particles suspended in water are selectively agglomerated by an oil or hydrocarbon liquid, and the agglomerates are recovered by screening. The process appears to be catalyzed by very small gas bubbles.

A series of four, geometrically similar mixing tanks was employed which ranged in volume from 0.346 to 11.07 liters. For an agglomeration test, a tank was fitted with a variable speed agitator which was equipped to indicate both speed and torque. During a batch agglomeration test agitator speed was held constant and the progress of agglomeration was monitored by observing changes in agitator torque in the case of concentrated suspensions or by observing changes in turbidity in the case of dilute suspensions. Tests were conducted with moderately hydrophobic coal from the Pittsburgh No. 8 Seam and with more highly hydrophobic coal from the Upper Freeport Seam using n-heptane, i-octane, and hexadecane as agglomerants.

Preliminary tests with dilute suspensions of Pittsburgh No. 8 coal showed that the process of agglomeration is triggered by the introduction of air bubbles and that the rate of agglomeration increases as more air is introduced. A subsequent study of the agglomeration kinetics of dilute suspensions showed that the rate of agglomeration is proportional to the particle number concentration raised to a power between 1.0 and 1.3. The rate increases with increasing amounts of either air or agglomerant and with increasing agitator speed. The rate is also greater for Upper Freeport coal than for Pittsburgh No. 8 coal, apparently because of the greater hydrophobicity of the former.

Agglomeration tests conducted with concentrated suspensions (e.g., 20 - 30 w/w% solids) produced changes in agitator torque which reflected the complex nature of the process. Several stages seem to be involved including gas dispersion, flocculation, coagulation, and agglomerate building. In some cases with higher agglomerant concentrations, small agglomerates combine to form large framboidal agglomerates. The minimum time t_e required to produce compact spherical agglomerates was found to be a key parameter which can be determined either by observing changes in agitator torque or by frequently sampling and examining a suspension undergoing agglomeration. The mean diameter d_p of the agglomerates produced by the end of each test was also determined along with the recovery of coal and its ash content.

Numerous agglomeration tests were conducted with two kinds of coal in concentrated suspensions using i-octane as an agglomerant in order to relate t_e and d_p to various system and operating parameters. In the case of Pittsburgh No. 8 coal, an excellent correlation was established between t_e and agitator power, system volume, and the concentrations of solids, i-octane, and air. A good correlation was also established between d_p and i-octane concentration, the ratio of impeller diameter to tank diameter, and agitator power input per unit volume. In addition, it was shown that the size of an agglomeration system for Pittsburgh coal can be scaled up by holding the group $V/P^{1.09}$ constant where V is the system volume and P is agitator power input. By holding this group constant a larger system should provide the same value of t_e as a smaller system.

Although good correlations for t_e and d_p with various parameters were also established for Upper Freeport coal, they were not the same as those obtained for Pittsburgh No. 8 coal. For example, in the case of Upper Freeport coal t_e did not appear to depend on solids concentration and d_p seemed to depend only on i-octane concentration. Also to scale up the size of an agglomeration system for Upper Freeport coal, the group $V/S^{5.43}$ should be held constant in order to maintain t_e constant. In other words, the impeller tip speed S is more important than agitator power in scaling up a system for Upper Freeport coal.

Most of the agglomeration tests conducted with concentrated suspensions of either Pittsburgh No. 8 coal or Upper Freeport coal resulted in a high recovery of an agglomerated product with a relatively low ash content. In many cases a coal recovery of 95% or more on a dry, ash-free basis was achieved together with an ash rejection of 70% or more. Coal recovery was observed to depend on agglomerate size which depends on other factors as already noted. In addition, coal recovery and ash rejection also depend on the efficiency of the procedure used for screening and washing the agglomerates. The procedure employed for this work was not necessarily the optimum since only a single screen size (i.e., 250 μm) was employed for product recovery and the method used for washing agglomerates was not investigated thoroughly.

INTRODUCTION

Several selective oil agglomeration processes have been proposed for cleaning fine-size coal in order to produce super clean coal with very low sulfur and ash contents (1-3). In these processes oil or a hydrocarbon liquid such as pentane or heptane is added to a vigorously agitated suspension of coal particles in water. The hydrophobic coal particles are selectively coated by the oil and stick together upon collision, whereas the hydrophilic mineral particles are generally unaffected. Since relatively large agglomerates are produced, they can be separated from the other materials by floating, skimming, or screening.

Although the basic concept for such a process had been known for many years, it had proved difficult to make the process cost effective and economically competitive. One reason for its relatively high cost had been an apparent need to use high-shear mixers which require a large power input (1,3). In order to provide high shear rates, it had become common practice to employ high-speed kitchen blenders for demonstrating and studying the process (2,4). Then it was discovered that the shear rate and power input could be greatly reduced by introducing some gas into a laboratory mixer which had been designed to exclude gas (5,6). This discovery was made while working with aqueous suspensions of ultrafine particles of a moderately hydrophobic coal from Illinois and using heptane as an agglomerant (5). When the suspensions were agitated at a moderate shear rate, only loose assemblages of particles or flocs were produced without gas present. However, when a small amount of air was present under similar conditions, agglomerates were produced which were compact and spherical.

Earlier work that led to this important discovery had indicated that the recovery of an Illinois coal by agglomeration with pentane or heptane was improved by having some air present in a modified kitchen blender (7). The blender had been modified so that the amount of air present could be controlled, but it still employed very high shear rates. Other evidence also suggested that agglomeration was promoted by the presence of a gas film on a coal surface (8,9). Moreover, the adsorption of a gas on the surface of a solid had been shown to make the surface hydrophobic (10), and it had also been shown that the more hydrophobic the coal, the greater its recovery with a given amount of oil in an agglomeration test (11).

What also needed to be demonstrated is that by incorporating gas in an agglomeration system, it would be possible to carry out the process of agglomeration in a standard type of industrial mixing system using moderate agitator power inputs and shear rates. One of the few reports of a study in which such a system had been used for agglomeration dealt with the agglomeration of chalk particles with hexane or kerosene in an aqueous suspension (12). The particles first had to be made hydrophobic by treatment with oleic acid. Agglomeration was conducted in both 10-liter and 100-liter mixing tanks of similar design. The smaller system consisted of a flat- bottom cylindrical tank and a variable speed agitator with a turbine impeller having vertical flat blades and a diameter equal to one-third that of the tank. The agitator power input could be varied over a range of 0 to 23 W/kg. The height and diameter of the tank were equal (23.6 cm), and the tank was fitted with four vertical baffles having a width equal to one-tenth of the tank diameter. Such a design is fairly standard for an industrial mixing system used for particle suspension or gas dispersion in a liquid (13). Although air may have been present in the mixing system used for agglomerating chalk, its importance was not recognized at the time so its effect was completely disregarded (12).

The present project was undertaken to provide a realistic, but laboratory-scale, demonstration of a gas-promoted oil agglomeration process for cleaning coal using scale model mixing systems representative of industrial practice. One objective was to determine the nature of the agglomeration mechanism when gas bubbles are present. A second objective was to measure the rate of agglomeration and to determine how the rate is affected by different system parameters. A third objective was to relate agglomerate size, yield, and quality to system and operating parameters. A fourth objective was to determine a suitable basis for size scale-up of the mixing system.

In order to meet these objectives, a series of four mixing tanks was constructed and utilized for several different series of agglomeration tests with two different coals. The tanks ranged in size from 0.346 to 11.07 liters and were geometrically similar to the 10 liter, flat bottom, cylindrical tank used by others for agglomerating chalk particles (12). The tanks were enclosed so that the amount of air present could be controlled from none to any specified amount. For an agglomeration test a tank was fitted with a variable speed agitator which was equipped to indicate both its speed and torque. In the case of dilute suspensions, the progress of agglomeration was monitored by observing changes in suspension turbidity. In the case of concentrated suspensions, progress was monitored by observing changes in agitator torque.

Several series of agglomeration tests were carried out with both dilute and concentrated suspensions of coal using pure hydrocarbon liquids such as heptane, i-octane, and hexadecane as “oils.” Finely ground samples of coal from both the Pittsburgh No. 8 Seam and Upper Freeport Seam were used for these tests. Dilute suspensions were employed initially to demonstrate the important role played by gas and to study the kinetics of agglomeration. Concentrated suspensions were employed later to determine the relationship between the time required to produce spherical agglomerates and various system and operating parameters or the relationship between agglomerate size and these parameters. Concentrated suspensions were also used for testing different system size scale-up rules and for determining the yield and quality of agglomerates produced. The percent recovery of coal on a dry, ash-free basis was taken as a measure of its yield, and the percent ash in the product as a measure of quality.

EXPERIMENTAL METHODS

Agglomeration Systems

Several scale model mixing systems were designed and constructed for conducting batch agglomeration tests. One of the principal components of each system was a flat-bottom, cylindrical tank which was fitted with a removable cover, four vertical baffles, and an agitator. Four tanks which differed in size were utilized. Since the principal dimensions of the tanks were kept in proportion, the tanks were geometrically similar (Table 1). However, the tanks differed with respect to minor construction details.

Figure 1 is a cross-sectional view of the tank which had an inside diameter of 15.24 cm (6.00 in.) and height of 15.75 cm (6.20 in.). The measured net volume of this tank was 2870 cm^3 (175 $in.^3$) when it was fitted with baffles and an agitator. Each of the four vertical baffles projected inward a distance of 1.27 cm (0.50 in.). The walls of the tank and baffles were made of Plexiglas, whereas the top and bottom were made of stainless

Table 1. Principal dimensions of mixing tanks.

	Diameter		Height		Baffle Width		Net Volume	
Tank No.	cm	in.	cm	in.	cm	in.	cm^3	$in.^3$
I	7.62	3.00	7.62	3.00	0.635	0.250	346	21
II	11.43	4.50	11.43	4.50	0.950	0.375	1,185	72
III	15.24	6.00	15.75	6.20	1.270	0.500	2,870	175
IV	24.00	9.45	24.13	9.50	1.900	0.750	11,071	676

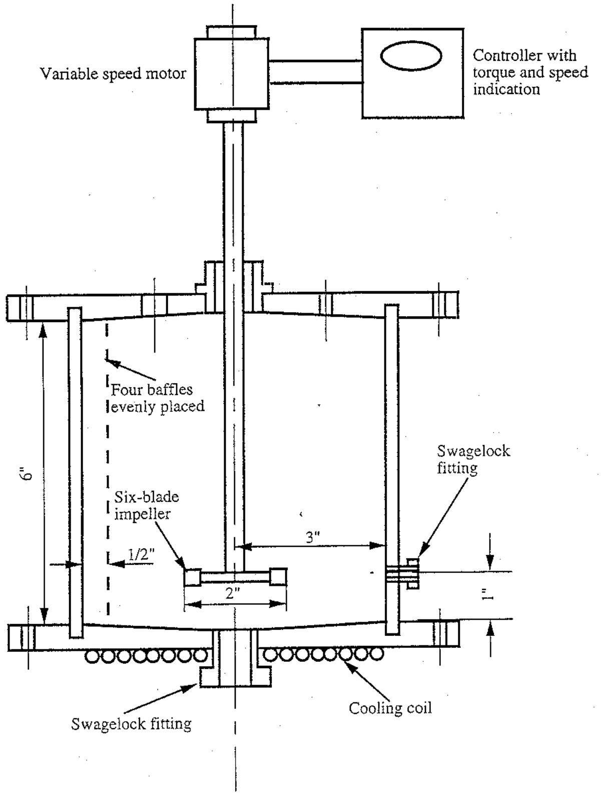


Figure 1. The model mixing system with the 15.24 cm (6.00 in.) diameter tank.

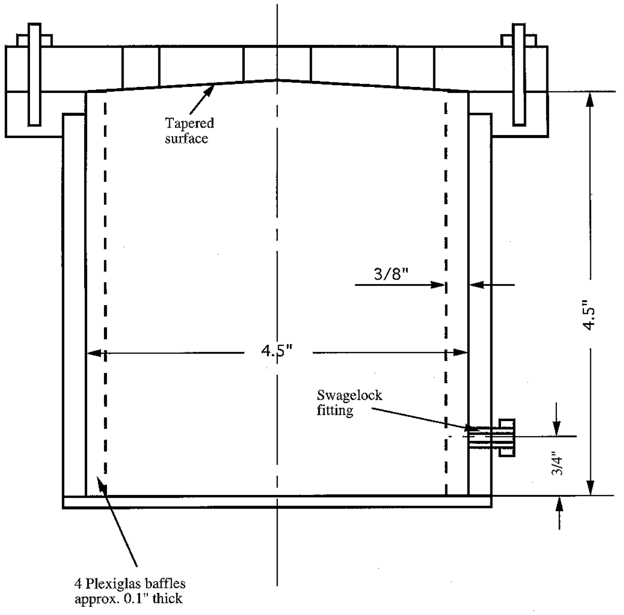


Figure 2. The 11.43 cm (4.50 in.) diameter mixing tank.

steel. The top and bottom were slightly concave to facilitate drainage of liquid from the bottom and venting of gas from the top. A cooling coil was attached to the bottom to remove heat generated by the agitator and thereby to control the temperature of the system. The top and bottom were easily removed to facilitate cleaning. Several openings were provided in the top of the tank for introducing material and for admitting the agitator shaft. An opening was provided in the bottom for draining the tank, and an opening was provided in the side for removing a stream of material needed for the measurement of turbidity. This opening was located 2.54 cm (1.00 in.) above the bottom.

Figure 2 is a cross-sectional view of a smaller tank that had an inside diameter of 11.43 cm (4.50 in.) and height of 11.43 cm (4.50 in.). The measured net volume of this tank was 1185 cm^3 (72.3 in.^3) when it was fitted with baffles and an agitator. The tank had four vertical baffles and each baffle projected inward a distance of 0.95 cm (0.375 in.). The tank and baffles were constructed completely of Plexiglas. The top was slightly concave to facilitate venting of gas, but the bottom was perfectly flat since it did not have a drain opening. Although the top could be removed easily, the bottom was attached permanently to the walls. Several openings in the top and one in the side were provided for the same reasons noted above for the larger tank. The side opening was located 1.91 cm (0.75 in.) above the bottom. To reduce the adhesion of oil-coated particles on the walls and other Plexiglas surfaces, the tank was treated with a mixture of Nochromix and concentrated sulfuric acid for 30 sec. and then was rinsed thoroughly with deionized water. This is a strongly oxidizing treatment which made the Plexiglas surface less oleophilic and more hydrophilic.

Two other tanks were constructed which were similar in design to the 11.43 cm diameter tank but lacked the side opening and neither tank received the Nochromix treatment. One of these tanks had an inside diameter of 7.62 cm (3.00 in.) and the other had an inside

Table 2. Principal dimensions of flat-blade turbine impellers

	Overall Dia.		Disk Dia.		Blade Length		Blade Height	
Impeller No.	cm	in.	cm	in.	cm	in.	cm^3	$in.^3$
I	3.65	1.44	2.40	0.94	0.85	0.33	0.64	0.25
II	5.08	2.00	3.30	1.30	1.25	0.49	1.00	0.39
III	6.35	2.50	4.16	1.64	1.50	0.59	1.29	0.51
IV	7.55	2.97	4.95	1.97	1.88	0.74	1.52	0.60

diameter of 24.0 cm (9.45 in.). Both tanks were made entirely of Plexiglas except for the bottom of the larger tank which was constructed from a thin sheet of stainless steel.

Agitation of the contents of each tank was provided by a single Rushton-type turbine impeller attached to a vertical drive shaft. This type of impeller has six vertical flat blades mounted on a horizontal disk. Four different impellers which were similar in design but differed in size were used interchangeably. The principal dimensions of these impellers are listed in Table 2.

For agglomeration tests involving dilute particle suspensions, the 15.24 cm diameter mixing tank was utilized together with the 5.08 cm diameter impeller which was located 2.54 cm (1.00 in.) above the tank bottom. For agglomeration tests involving concentrated particle suspensions, all four sizes of tanks and impellers were used. In each case the impeller was located midway between the top and bottom of the tank.

Three variable speed agitator drive systems were available for use with the mixing tanks. The power output ratings of these systems was as follows: 41 watts (1/18 hp), 93 watts (1/8 hp), and 186 watts (1/4 hp). The largest system was reserved for the largest mixing tank. The smallest system was used for runs with dilute suspensions, while the intermediate size system was used for concentrated suspensions and for some dilute suspensions. Each of these systems was provided with both a speed indicator and a torque indicator. The calibration of each of these indicators was checked. Although the largest drive system was equipped with an accurate torque indicator, the two smaller systems were not and calibration curves had to be developed for use with these systems. Each of the drive systems could be operated over a wide range of speed. The maximum operating speed was 2000 rpm for the largest system and 2500 rpm for the others.

For monitoring the turbidity of dilute suspensions undergoing agglomeration, the 15.24 cm (6.0 in) diameter tank was used together with a photometric dispersion analyzer (PDA) for measuring the turbidity. A PDA 2000 instrument manufactured by Rank Brothers Ltd. was used for this purpose. During an agglomeration test, a stream of material was drawn continuously from the mixing tank and conducted through the measuring cell of the PDA where the light transmittance of the suspension was determined. After passing through the measuring cell, the suspension was returned to the mixing tank by means of a peristaltic pump.

Oil Agglomeration Materials and Methods

A large number of oil agglomeration experiments were conducted with Pittsburgh No. 8 Seam coal from Belmont County, Ohio, and with Upper Freeport Seam coal from Indiana County, Pennsylvania. The Pittsburgh No. 8 coal is regarded as high volatile A bituminous coal whereas the Upper Freeport coal is probably a medium volatile bituminous coal. After drying the coal, the ash and sulfur contents of the Pittsburgh coal were 28 wt.% and 5.0 wt.%, respectively. Two different batches of Upper Freeport coal were utilized. The ash and total sulfur contents of the first batch were 14 wt.% and 2.0 wt.%, respectively, while the ash content of the second batch was 26.5 wt.%, all on a dry basis.

Coal particles were suspended in deionized water having a resistivity of 17-18 megohm·cm for agglomeration tests. For many of these tests the agglomerant was either n-heptane or i-octane obtained from Burdick and Jackson Laboratories, Inc. According to the supplier, these materials had been distilled in glass with the result that the normal boiling point range was 98-99°C for the n-heptane and 99-100°C for the i-octane, which indicates a high degree of purity. For some of the later tests with concentrated suspensions, the agglomerant was pesticide grade i-octane from Fisher Scientific with an indicated purity of 99.5%. For a few tests the agglomerant was either hexadecane (99%) with a normal boiling point of 287°C or squalane (99%) with a boiling point of 176°C at a pressure of 0.05 mm Hg. These materials were obtained from Aldrich Chemical.

To prepare coal for agglomeration tests, the material was first crushed with a jaw crusher and then either ground with a roll mill or a high-speed impact mill. The material was ground further with a stirred ball mill to produce particles having an average size of about 10 ìm or less. For this step the ball mill was operated for 20 min. with a charge consisting of 250 g coal, 250 g water, and 1200 g stainless steel balls having a diameter of 3 mm. A stirring speed of 540 rpm was employed. The product was partially dewatered with a Büchner filter funnel and then stored as a paste containing approximately 56% solids. The paste was stored either under an atmosphere of argon or in a refrigerator set at 5°C to minimize oxidation.

To prepare for an agglomeration test, one of mixing tanks was partly filled with deionized water and a measured quantity of coal paste was added and dispersed in the water by gentle stirring. For some experiments the coal suspension was then degassed by applying a partial vacuum to the partly filled mixing system at room temperature for 20 min. The partial vacuum corresponded to an absolute pressure of 5-6 kPa. Afterwards the mixing system was topped off with degassed water, and any remaining gas bubbles were removed from the system.

The procedures for conducting agglomeration tests differed for dilute and concentrated particle suspensions because different monitoring techniques were used. In the case of dilute suspensions, the progress of agglomeration was followed by observing changes in the turbidity of a suspension, whereas in the case of concentrated suspensions agglomeration was tracked by observing changes in agitator torque.

In the case of dilute suspensions, some agglomeration tests were performed by introducing air before oil, while for other tests oil was introduced before air. Two different methods were used for introducing air. When the volume of air was small (e.g., 2.5 ml), the air was injected near the impeller by using a syringe with a long needle. When the volume was larger, air was introduced by withdrawing a measured volume of slurry which was replaced by an equal volume of air.

For dilute suspensions when air was introduced before oil, the coal suspension was agitated for 3 min. at a given speed after air was introduced. A measured quantity of oil was then injected with a syringe, usually in a single dose. As agitation was continued at the same rate, the progress of agglomeration was monitored by observing the change in turbidity of the suspension. As the particles combined to form agglomerates, the particle concentration decreased which reduced the turbidity of the suspension. Since the reduction in turbidity was reflected by an increase in the output signal from the photometric dispersion analyzer, the rate of increase of the output signal was indicative of the apparent rate of agglomeration.

When oil was introduced before air, a dilute coal suspension was stirred for 9 min. at a given speed after oil was introduced. A measured quantity of air was then added and the test was continued as described above.

In the case of dilute suspensions, no attempt was made to control the temperature of the system during an agglomeration test. At the start of a test, the system was at room temperature, and as the test proceeded the temperature rose gradually due to agitator power input. For a 5.08 cm diameter impeller operating in a 15.24 cm diameter tank, the temperature of the system increased 2.5°C/hr at 1500 rpm and 13°C/hr at 2400 rpm.

For conducting an agglomeration test with a concentrated suspension, one of the mixing tanks was charged with deionized water and a measured amount of coal paste was dispersed in the water by gentle stirring. The quantity of slurry was chosen to occupy approximately 80% of the tank volume. For some experiments the slurry was degassed next by connecting the tank to a vacuum pump and applying a vacuum corresponding to –95 kPa for 20 min. During this time the tank was alternately shaken and tapped against the laboratory bench to release air bubbles from the slurry. The tank was opened subsequently and a measured amount of i-octane was introduced. The agitator was inserted, and the tank was closed. Next the tank was filled completely with deionized water which had been degassed by the above procedure. Great care was taken to insure that no gas bubbles were present in the system. With the temperature of the system close to room temperature (19°C), the agitator was turned on and operated at a preselected speed. After 5 min. of stirring and conditioning a measured volume of slurry was withdrawn from the tank and a corresponding volume of air was admitted. This operation required about 30 s. After air was introduced the test was continued. During a test the agitator was operated at a constant speed, and the indicated torque was recorded. The temperature of the system was kept between 19 and 21°C by circulating cooling water through a coil attached to the bottom of the 15.24 cm diameter tank or by surrounding the other tanks with an ice bath. Small samples (1-2 cm^3) of the coal suspension were withdrawn from the system at various time intervals by using a syringe equipped with a needle having a large bore (1.5 mm). As each sample was removed, a corresponding amount of water was added so that the volume of air in the system remained constant. After a test was completed, the samples were examined with an optical microscope to determine the general shape and size of particle aggregations present at various stages of agglomeration. Image analysis was used to determine the size distribution of the product during the later stages of the project.

Although the system was degassed initially for many of the earlier agglomeration tests with concentrated suspensions, this step was omitted for most of the later tests. Except for vacuum degassing, the experimental procedure for conducting a test was the same.

In some of the preliminary tests with concentrated suspensions the focus was on determining the size and shape of the agglomerates and the time required to produce compact, spherical agglomerates so no attempt was made to determine the yield and purity of the agglomerates. In later tests the agglomerates were recovered by screening the final suspension. First this was accomplished by diluting the suspension with about an equal volume of water and pouring the mixture onto a 250 ìm screen. Later the procedure was modified. The contents of the mixing tank were dumped into a deep vessel fitted with 250 ìm screen bottom which was placed in a pail of water. After the agglomerates had been washed, they were recovered, dried, weighed, and analyzed for their ash content. The tailings left in the pail were recovered by filtration and then dried, weighed, and analyzed.

RESULTS ACHIEVED WITH DILUTE SUSPENSIONS

Since the turbidity of a particle suspension is proportional to the number concentration and scattering cross section of the various particles present, its measurement provides a convenient means for monitoring the agglomeration of dilute suspensions which undergo a large drop in particle concentration as agglomeration takes place. Turbidity is measured by determining the attenuation of a narrow beam of light as it passes through the suspension. The following relation based on the Beer-Lambert law is employed for determining the turbidity τ:

$$\tau = (1/L)\ \ln\ (I_o/I) \qquad (1)$$

I represents the measured intensity of a narrow beam of light after passing a distance L through the suspension, while I_o represents the intensity of a similar beam of light which has traveled the same distance through clear water.

For the present investigation, the turbidity of a suspension was measured with a photoelectric dispersion analyzer (PDA) as the suspension flowed through a glass tube having an inside diameter of 2.5 - 3.0 mm. Since the output signal voltage V from the PDA was proportional to the intensity of the beam of light after passage through the tube, the preceding relationship was used in the following form:

$$\tau = (1/L)\ \ln\ (V_o/V) \qquad (2)$$

During a batch agglomeration test the turbidity decreased and the output voltage V increased. Therefore, the output voltage was a good indicator of agglomeration.

Several series of experiments were conducted using the measurement of turbidity to study particle agglomeration in dilute suspensions. One series of tests was used to define the role of air in the oil agglomeration process. A second series was used to demonstrate the apparent agglomeration of hydrophobic particles by air bridges. A third series was used for measuring the rate of agglomeration of coal particles and for developing an understanding of the kinetics of gas-promoted oil agglomeration.

The Role of Air in Oil Agglomeration

The first series of batch agglomeration tests with a dilute suspension was conducted with finely ground Pittsburgh No. 8 coal using 20 v/w% heptane (0.20 ml heptane/g coal) as an agglomerant. These tests were conducted with the 15.24 cm diameter mixing tank and an agitator which was fitted with a 5.08 cm diameter impeller mounted 2.54 cm above the bottom of the tank. The net volume of the mixing tank was 2870 ml. The progress of agglomeration was observed by monitoring the output signal from the PDA.

Results. For the first set of three tests, the previously prepared coal paste was suspended in degassed water. For each test the suspension was stirred at 1500 rpm for 5 min. before introducing heptane into the “closed” system. After adding heptane, stirring was continued at the same rate, and until air was introduced the output voltage of the PDA remained constant indicating no agglomeration. However, as soon as air was introduced, the output voltage climbed rapidly showing that agglomeration was taking place. This phenomenon was observed in all three tests which differed only in the length of time between the point of heptane introduction and air introduction (see Figure 3). Therefore, it was apparent that agglomeration was triggered by the introduction of air.

The necessity of having a gas present was further illustrated by the next set of agglomeration tests. For these tests the finely ground coal was also suspended in degassed water. In each test after heptane was introduced, the suspension was stirred for 9 min. before a measured quantity of air was added. The amount of air was varied among tests,

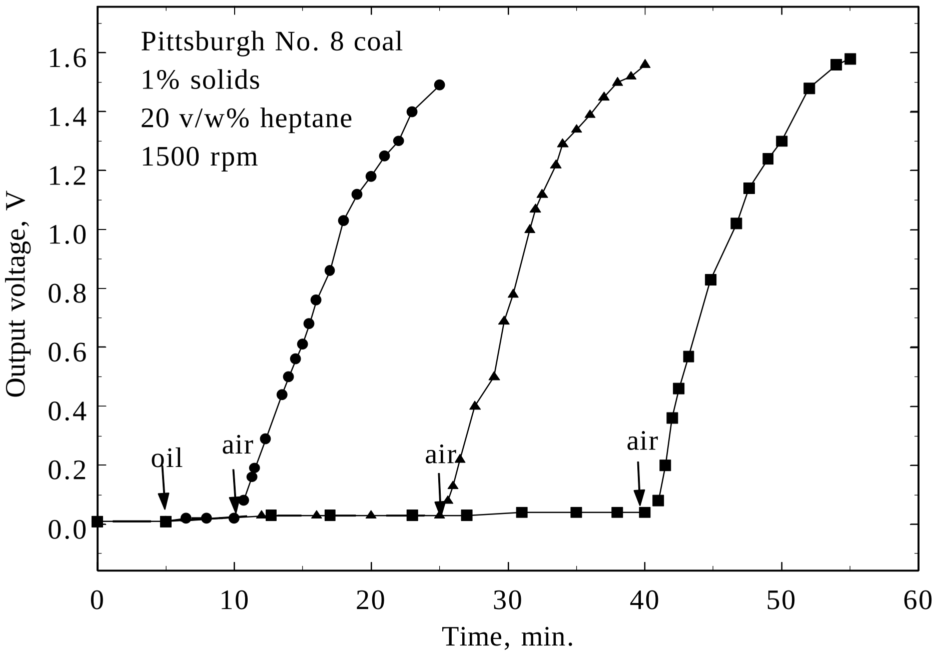


Figure 3. Results of three agglomeration tests in which 50 cm^3 of air was added at either 5, 20, or 35 min. after introducing heptane.

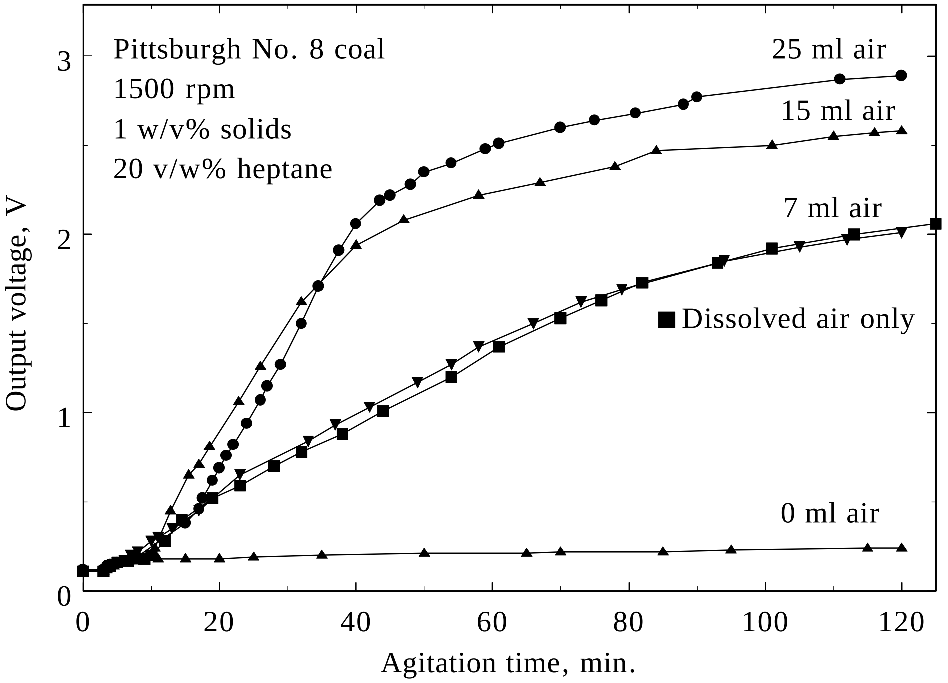


Figure 4. Results of agglomeration tests with Pittsburgh coal suspended in water which had been degassed followed by the introduction of different amounts of air.

whereas the following parameters were kept constant: 1.00 w/v% solids, 20 v/w% heptane, and 1500 rpm stirring speed. The results presented in Figure 4 indicate that when no air was added to the mixing system, the output signal from the PDA remained flat for up to 2 hr showing that no agglomeration occurred. However, when only 7 ml of air was added 9 min after heptane was introduced, the output signal immediately started to rise and continued to rise steadily showing that agglomeration was taking place. The addition of larger amounts of air produced even larger and more rapid increases in the output signal indicating higher rates of agglomeration. Interestingly, when another experiment was conducted by agglomerating coal in water which had not been degassed, the results, which are also reported in Figure 4, were similar to those achieved when 7 ml air was added to the degassed system. Therefore, it appeared that air dissolved in the water was sufficient to promote agglomeration. But did the air have to come out of solution to promote agglomeration?

To answer this question additional experiments were conducted using water which had not been degassed and hexadecane and squalane as agglomerants. No air was added to the system other than the air dissolved in the water. The results achieved with the different agglomerants and shown in Figure 5 indicate that the rate of agglomeration with either hexadecane or squalane was much smaller than with heptane. Although the solubility of heptane in water is small, it is still appreciably greater than that of hexadecane or squalane. The results suggest that when heptane is used as an agglomerant some heptane dissolves in the water and displaces enough air from the water to promote agglomeration. Hexadecane and squalane being much less soluble displace less air, and, therefore, have much less effect. This hypothesis was tested further by first saturating water with heptane and then using the water to suspend coal for a test of agglomeration. The water was not degassed

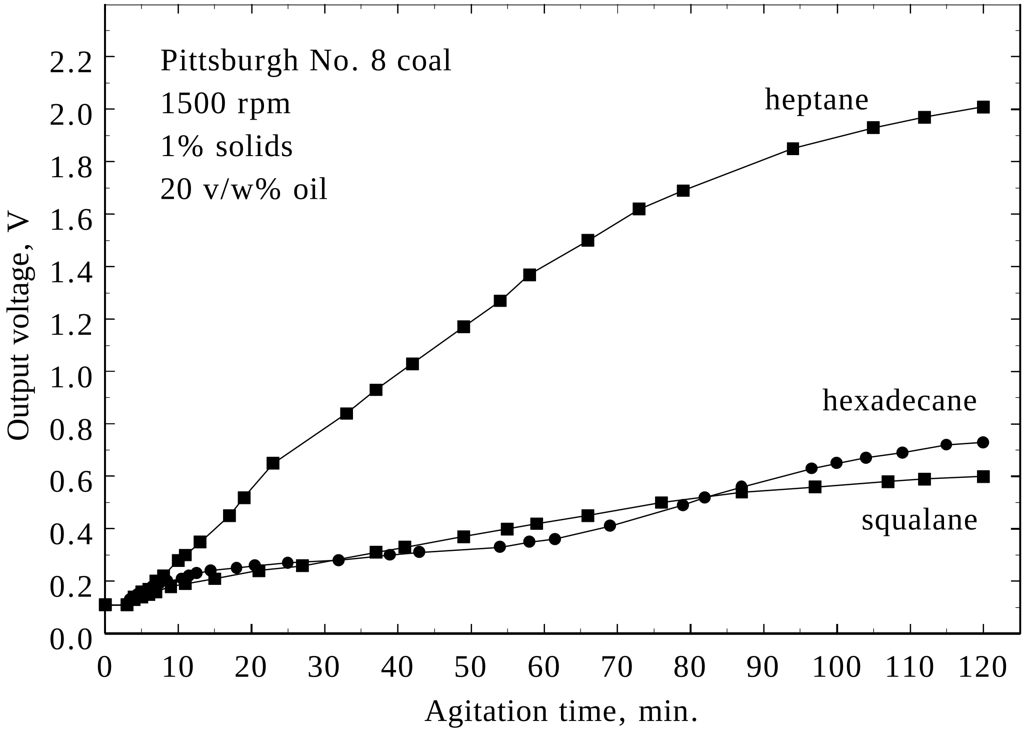


Figure 5. Results of agglomeration tests with Pittsburgh coal suspended in water which had not been degassed.

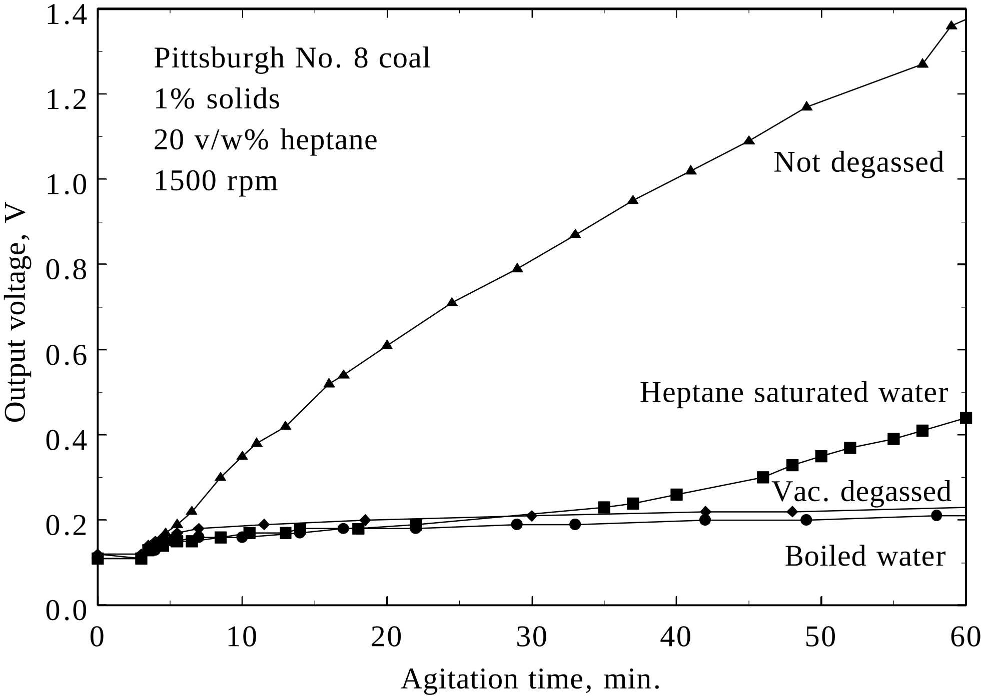


Figure 6. Results of agglomeration tests with Pittsburgh coal suspended in water which had received different treatments.

otherwise and any excess heptane was removed by evaporation at room temperature before conducting the agglomeration test. The results of the agglomeration test which are reported in Figure 6 show that virtually no agglomeration took place during the first 35 min, and then agglomeration proceeded very slowly. The results were in sharp contrast to those observed when an agglomeration test was conducted using water which had not been degassed. Also the results obtained with heptane saturated water were very similar to those observed with water which had been degassed by applying a partial vacuum. Therefore, it seems very likely that the displacement of air from water by heptane is sufficient to promote the agglomeration of Pittsburgh No. 8 coal.

Another method of degassing water used for agglomeration was also tested. This method involved boiling the water for 30 min to remove dissolved air. The water was then cooled rather quickly and used immediately for an agglomeration test. The results of this test which are shown in Figure 6 were almost identical to the results which had been obtained by vacuum degassing.

While the preceding experiments showed that even small amounts of air can promote the oil agglomeration of Pittsburgh No. 8 coal, the air had to be present as a separate phase. Apparently heptane is capable of displacing enough air dissolved in water at normal temperature and pressure to promote agglomeration. To test this theory additional experiments were conducted using water which had not been degassed. Furthermore, no air was introduced during the experiments.

The results of one set of experiments are indicated by Figure 7. In this set the solids concentration was varied among experiments while the heptane concentration and stirring speed were kept constant. The results indicate that the material was agglomerated in each case, and since the PDA output signal increased more slowly as the particle concentration

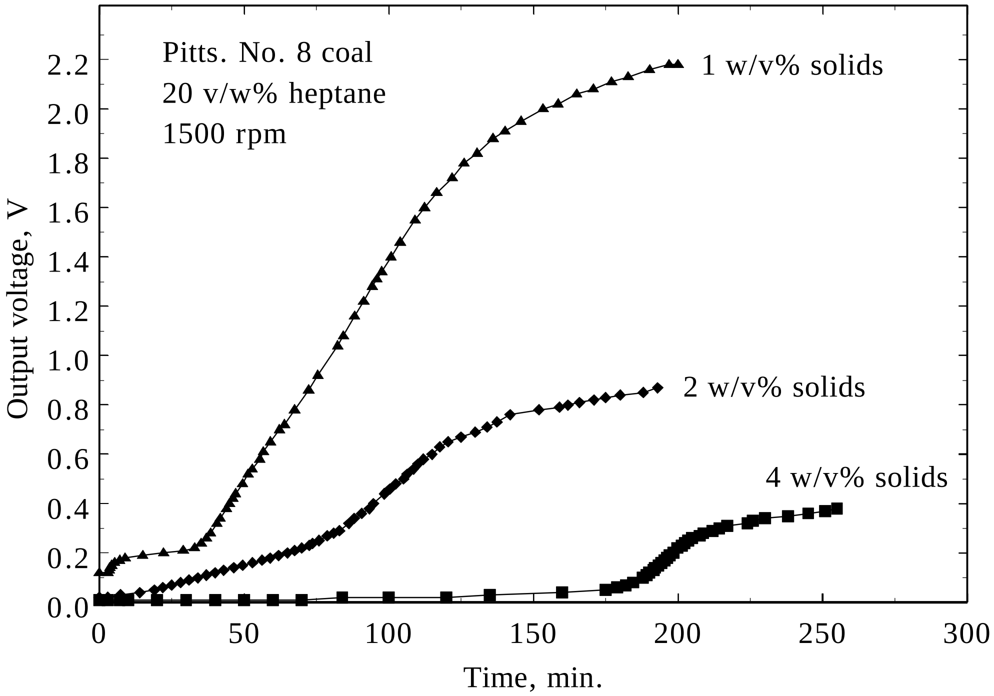


Figure 7. Effect of solids concentration on agglomeration of Pittsburgh coal suspended in water which had not been degassed.

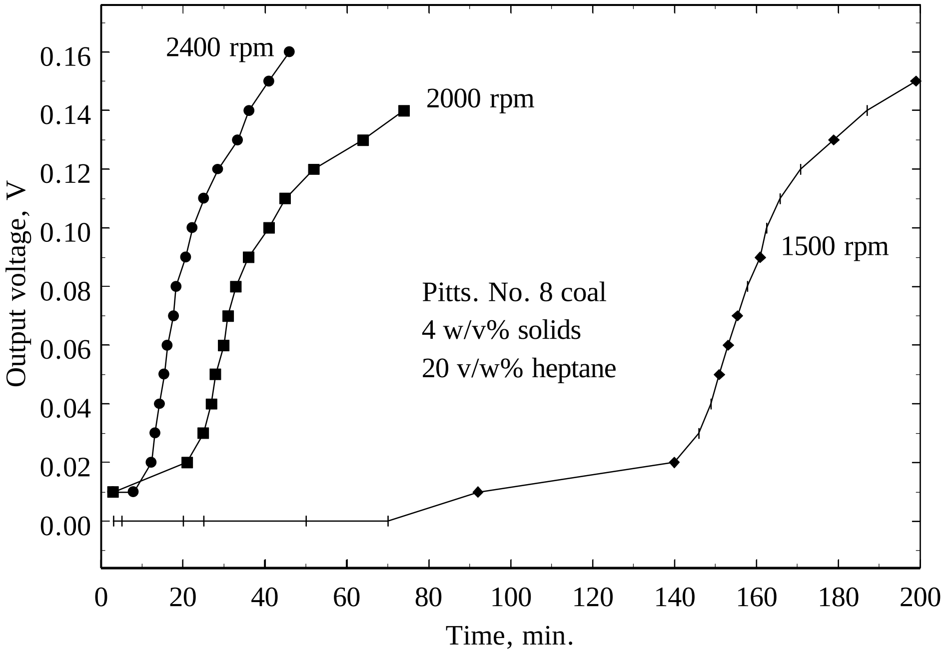


Figure 8. Effect of agitator speed on agglomeration of Pittsburgh coal suspended in water which had not been degassed.

was raised, it appeared that the rate of agglomeration decreased with increasing particle concentration. However, these results are deceptive because many more particles had to be agglomerated to achieve the same level of turbidity when starting with a higher initial particle concentration.

The results of another set of experiments in which no air was added are shown in Figure 8. In this set the stirring speed was varied while the solids concentration and heptane concentration were kept constant. Again it appeared that agglomeration was promoted by air which was initially dissolved in the water. Moreover the rate of agglomeration increased with stirring speed, and the increase in rate was particularly striking when the speed was increased from 1500 rpm to 2000 rpm. While an increase in the rate of agglomeration with increasing stirring speed was anticipated, the unexpected large increase caused by raising the speed from 1500 rpm to 2000 rpm called for an explanation. One possibility is that a small amount of air was drawn into the system at the higher speeds. Another possibility is that impeller cavitation appeared at the higher speeds and promoted the oil agglomeration process.

Since air seemed to play a key role in the agglomeration process, further experiments were conducted to study the effect of air concentration on the apparent rate of agglomeration. For these experiments finely ground coal was suspended in water which had not been degassed, and in most cases a measured amount of air was added to the system initially. After stirring the suspension for 3 min., a measured volume of heptane was introduced and agglomeration was observed by following the PDA output signal. The results of agglomeration tests are shown in Figure 9 for a solids concentration of 1.00 w/v% and in Figure 10 for a solids concentration of 4.00 w/v%, respectively. For both solids concentrations the rate of agglomeration increased with increasing amounts of air. However,

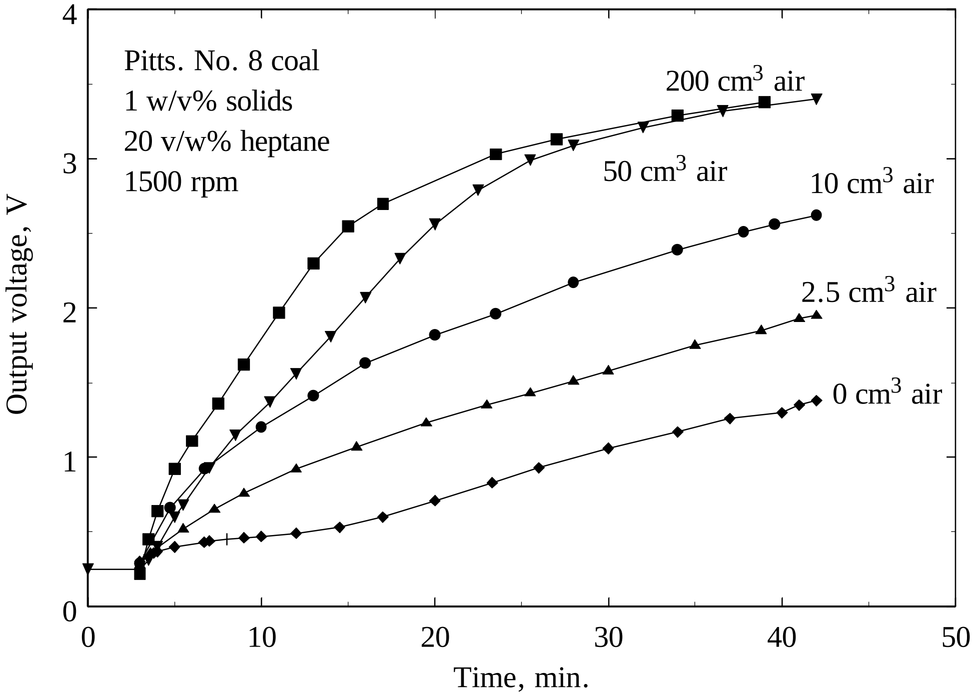


Figure 9. Results of agglomeration tests with Pittsburgh coal suspended in water which had not been degassed followed by the introduction of the indicated amount of air.

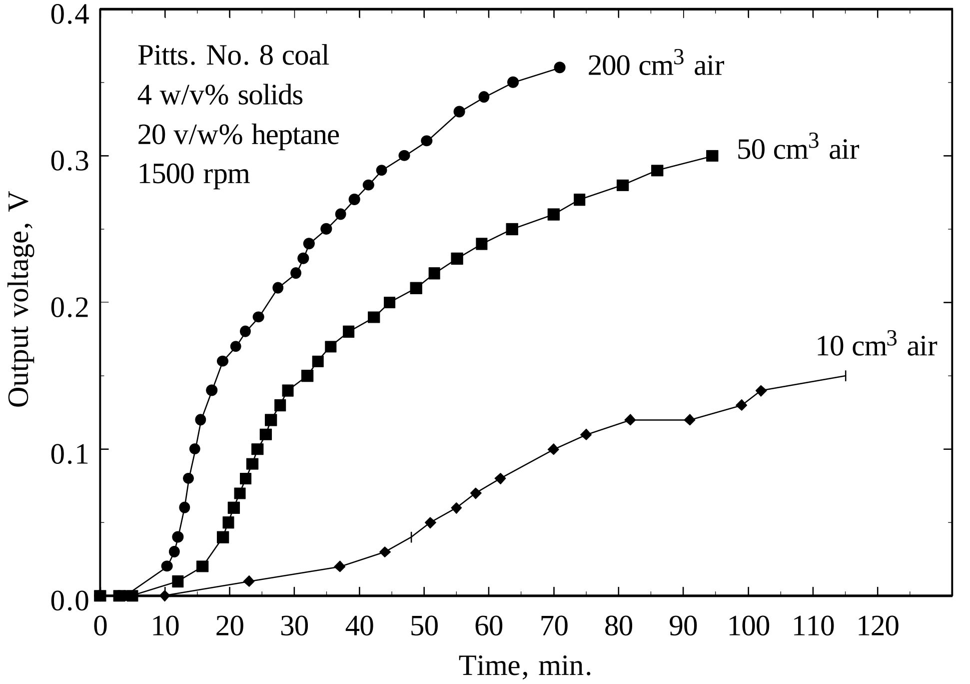


Figure 10. Results of agglomeration tests with a higher concentration of Pittsburgh coal suspended in water which had not been degassed followed by the introduction of the indicated amount of air.

the effect of air concentration appeared to be nonlinear so that the rate of increase in agglomeration rate with respect to air concentration seemed to decline as the concentration was raised.

Other experiments were conducted with added air and different particle concentrations, and the results are shown in Figure 11. A comparison of these results with those presented in Figure 7 shows that for any given solids concentration the rate of agglomeration was much greater when 50 ml of air was added to the system than when the only air present was that initially dissolved in the water in which the coal was suspended.

Other experiments were conducted with added air to show the effect of agitator speed. One set of experiments was conducted with a solids concentration of 1.00 w/v% (see Figure 12) and another set with a solids concentration of 4.00 w/v% (see Figure 13). The results indicate that for an agitator speed of 1250 rpm or more the rate of agglomeration was relatively high and that the rate of agglomeration increased with increasing stirring rate. Figure 12 also indicates that the greatest increase in agglomeration rate was produced by an increased in agitator speed from 1000 to 1500 rpm. Further increases in agitator speed produced progressively smaller increases in agglomeration rate so that an increase in speed from 2000 rpm to 2400 rpm caused only a slight increase in agglomeration rate. In addition, Figure 12 indicates that for an initial solids concentration of 1.00 w/v%, the rate of agglomeration at 1000 rpm was very slow, and Figure 13 suggests that for an initial solids concentration of 4.00 w/v% the rate of agglomeration at 1000 rpm was unobservable.

A comparison of the results in Figure 13 with those in Figure 8 shows that with an agitator speed of either 1500 rpm or 2400 rpm the rate of agglomeration was much greater with 50 ml of added air than with no added air.

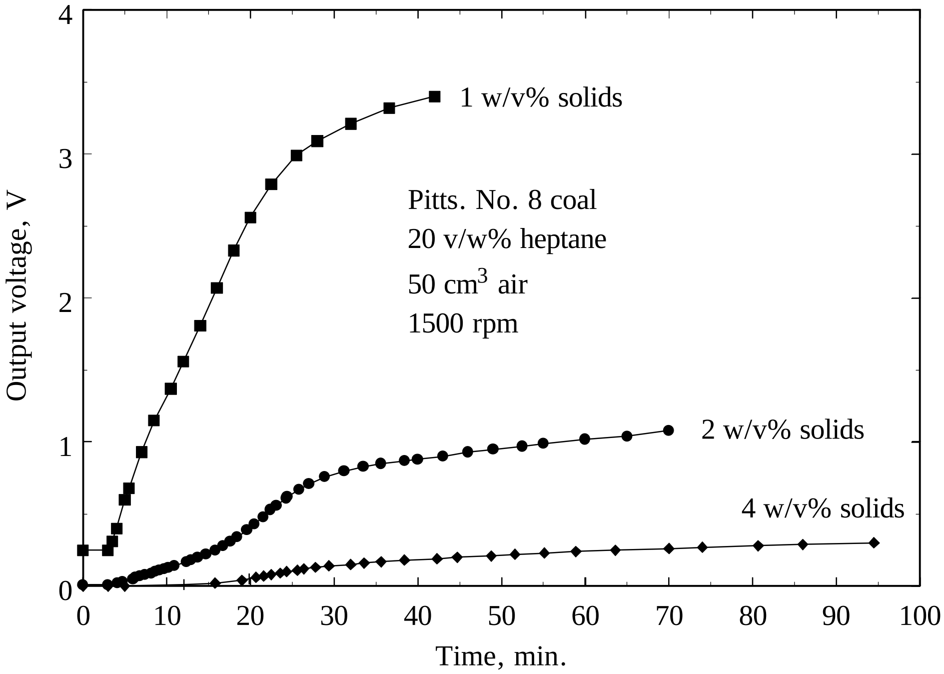


Figure 11. Agglomeration of small concentrations of Pittsburgh coal suspended in water which had not been degassed plus 50 cm^3 of air added.

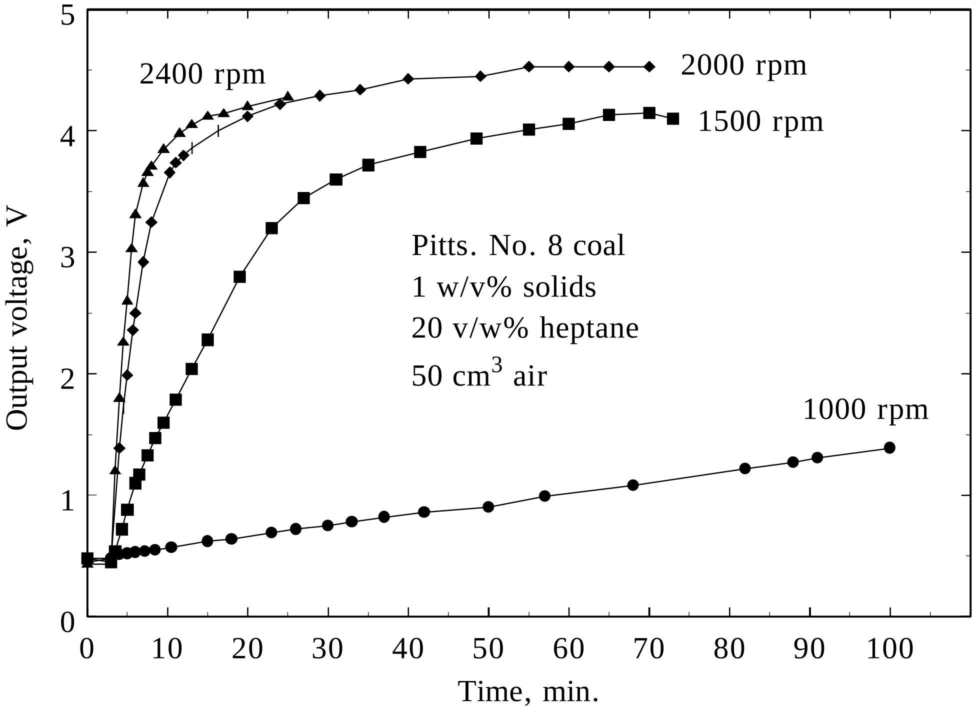


Figure 12. Effect of agitator speed on agglomeration of 1 w/v% concentration of Pittsburgh coal suspended in water which had not been degassed and which had 50 cm^3 of air added.

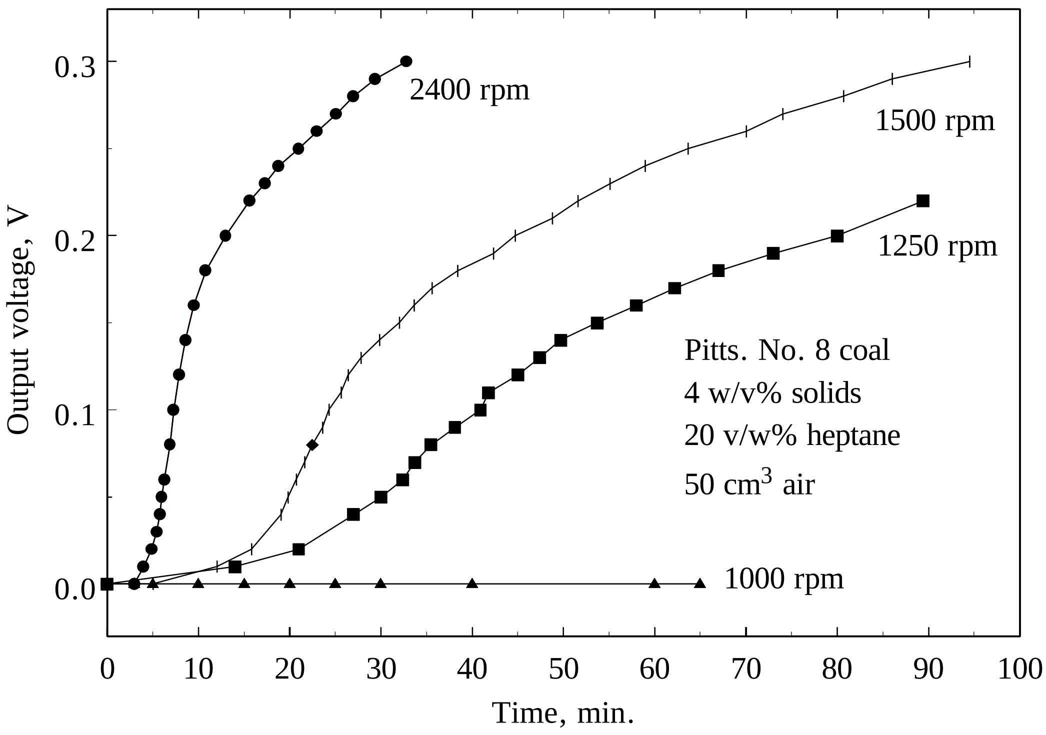


Figure 13. Effect of agitator speed on agglomeration of 4 w/v% concentration of Pittsburgh coal suspended in water which had not been degassed and which had 50 cm^3 of air added.

Discussion of the Role of Air. The preceding results show that the process of agglomeration of a moderately hydrophobic coal with heptane in an aqueous suspension is triggered by a small amount of air present as a separate phase. The quantity of air required to trigger agglomeration depends on solids concentration and the rate of agitation. More air is required for a higher solids concentration than for a lower concentration, and less air is required with a higher agitator speed than for a lower speed. The rate of agglomeration also depends on the quantity of air present as well as agitator speed.

Although the nature of the role played by air in oil agglomeration is not completely clear, several possibilities come to mind. One possible role is that air increases the apparent hydrophobicity of coal by occupying surface cracks, crevices, and pore mouths (6). Since air is much more hydrophobic than coal, the net effect is to create a surface which on average is more hydrophobic than the solid. The effect of an air film on the wetting characteristics of a higher rank bituminous coal became readily apparent when an attempt was made to mix the dry powder with water. An almost impossible task was easily accomplished by first degassing the mixture of powder and liquid.

A second role which air may play in coal particle agglomeration could arise from the adsorption of submicroscopic gas nuclei or tiny gas bubbles on the surface of the coal. It has been shown that micro-bubbles are generated by cavitation produced by high-shear mixers, and the process is favored by a high dissolved gas content (14). Particle bridging by submicroscopic bubbles may have accounted for the hydrophobic coagulation of coal particles observed by Xu and Yoon (15). This possibility seems likely in view of the evidence presented by Parker and Claesson (16) which showed that the long-range force of attraction between hydrophobic surfaces is due to bridging by submicroscopic gas bubbles. Furthermore, it has been proposed to improve the froth flotation process used for separating minerals by first coating the mineral particles to be floated with gas nuclei, since this will accelerate the subsequent attachment of the particles to macro-bubbles and thereby improve the flotation rate (14). Therefore, gas bubbles may promote oil agglomeration either by making the surface of coal more hydrophobic or by serving to bridge particles.

There may be an additional role for gas bubbles to play in oil agglomeration because of the observed interaction of particle flocs and flakes with larger gas bubbles at an intermediate stage of the process before spherical agglomerates are produced (17). Since the final agglomerates are not associated with these bubbles, it appears as though the bubbles serve as a “catalyst” for the formation of the agglomerates.

Air Agglomeration of Various Hydrophobic Solids

While attempting to wet and disperse dry particles of highly hydrophobic materials in water, it was discovered that the process of wetting was facilitated greatly by vacuum degassing of the mixture of particles and water. Therefore, it was reasoned that once dispersed in water, such particles would have a tendency to collect at an air-water interface, if a gas were dispersed in the liquid, which could in turn lead to the formation of air bridges between particles similar to the oil bridges between particles in oil agglomeration.

To investigate this concept a series of experiments was carried out in which aqueous suspensions of various hydrophobic solids were prepared by degassing and agitation, and then air was introduced in a series of small increments into the agitated suspensions. Evidence of particle agglomeration was obtained by observing the change in turbidity of a given suspension and by examining the product with an optical microscope.

Hydrophobic Materials. The hydrophobic solids included Teflon powder, flowers of sulfur, and finely ground gilsonite, Ceylon graphite, and Pittsburgh No. 8 coal. The source, type of preparation, and particle size of each material are indicated in Table 3. The Teflon powder was designated by the manufacturer as No. 8 compression molding resin, and it consisted of potato-shaped pellets which ranged from 0.1 to 1.0 mm in size.

The hydrophobicity of the different materials was evaluated by measuring the three-phase, air/water/solid contact angle using the captive bubble measurement technique. A small air bubble was brought in contact with a flat, polished surface of the material under water, and both the water advancing and water receding contact angles were measured through the water phase with a goniometer. In the case of Teflon, a piece of flat sheet was used for the measurement. It was first cleaned with Nochromix and then rinsed with deionized water but was not polished. The results of these measurements are shown in Table 4. Since Teflon

Table 3. Materials used for air agglomeration

Material	Source	Preparation	Size, mm
Teflon	Dupont Co.	None	0.575
Gilsonite	Uinta Basin	Dry grinding	<0.180
Sulfur	Fisher Scientific	None	<0.038
Graphite	Ceylon	Dry grinding	<0.038
Coal*	Pitts. No. 8 Seam	Wet grinding	0.010

*hvAB, 27% ash, 5% S

Table 4. The three-phase contact angle of different materials

Material	Contact Angle, degrees			Remarks
	θ_A	θ_R	θ_{AV}	
Teflon	109°	86°	98°	Cleaned/Not polished
Gilsonite	89°	43°	66°	Wet Polished
Sulfur	85°	45°	65°	Wet Polished
Graphite	81°	42°	61°	Dry Polished
Graphite	13°	13°	13°	Wet Polished
Pitts. Coal	58°	25°	41°	Wet Polished
Pitts. Coal	0	0	0	Slime Coated

θ_A is the advancing contact angle

θ_R is the receding contact angle exhibited the largest contact angle, it was the most hydrophobic material and, therefore, the most difficult to wet. The hydrophobicity of graphite was found to depend on whether it was prepared by dry polishing or wet polishing. The dry polished material may have been more hydrophobic than the wet polished material because of the presence of air in pores, cracks, and crevices. Although clean, wet polished Pittsburgh No. 8 coal was moderately hydrophobic with an average contact angle of 41°, it became completely hydrophilic when the polished surface was exposed first to a suspension of finely ground coal which apparently left a slime coating on the surface.

Agglomeration Tests. To prepare a suspension for agglomeration, a measured quantity of dry particles and 2800 cm^3 of water were placed in the 15.24 cm (6.00 in.) diameter mixing tank described above. The tank was sealed and connected to a vacuum pump. The suspension was degassed by applying a vacuum corresponding to -95 kPa for 20 min., and then the tank was topped off with previously degassed water. In most cases the suspension containing 1-2% solids was conditioned for 2 min. by operating the agitator at 2400 rpm. As agitation was continued, air was introduced in a series of measured increments, and the progress of agglomeration was monitored by observing changes in the turbidity of the system. For coal to be agglomerated, it was first necessary to treat the material with a small amount of heptane. The suspension was then conditioned for 5 min. before proceeding with the test. The relative turbidity change ($\Delta\tau_r$) in percent defined below was used as a measure of the extent of particle agglomeration.

$$\Delta\tau_r = \left[(\tau_o - \tau)/\tau_o\right]100 \qquad (3)$$

In this expression τ_o represents the initial turbidity of the unagglomerated suspension and τ represents the turbidity after agglomeration has taken place. It is apparent that as

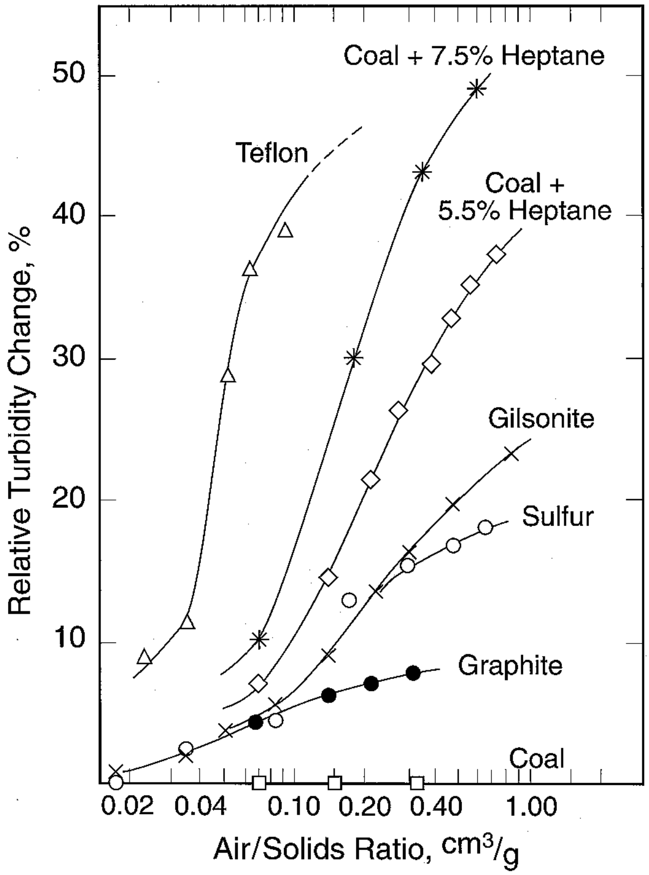


Figure 14. Results of air agglomeration tests of different hydrophobic materials.

agglomeration takes place the relative turbidity change will increase while the absolute turbidity decreases.

The results of the series of agglomeration experiments conducted with different hydrophobic solids are shown in Figure 14. It can be seen that for each material except untreated Pittsburgh No. 8 coal the relative turbidity change of the corresponding suspension increased as more air was introduced. Therefore, except for the untreated coal, air promoted the agglomeration of the various materials. Apparently the hydrophobicity of the untreated coal was insufficient to cause agglomeration. However, it was found that by preconditioning the coal with a small amount of heptane, the material responded to air agglomeration in the same way as the other hydrophobic solids responded. The slope of each line plotted in Figure

14 seemed to be proportional to the hydrophobicity of the corresponding material as indicated by its three-phase contact angle.

When samples of the Teflon particle suspension which had been treated with air were examined with an optical microscope, numerous particle aggregates or agglomerates were observed. The nature of the particle bonding was not apparent because there were no obvious air bubbles or air bridges between particles. However, when the sample was warmed gently by the light applied to view the sample with the microscope, air was expelled from within the agglomerates causing gas bubbles to appear on the surface of the agglomerates.

The treatment of gilsonite particles, which were considerably smaller and less hydrophobic than the Teflon particles, produced aggregates of particles which differed from those observed with Teflon. The gilsonite particles either tended to coat the surface and stabilize the somewhat larger gas bubbles or to form small and rather loose clusters or flocs which often appeared to be associated with smaller gas bubbles.

The treatment of even less hydrophobic materials such as graphite did not produce particle aggregations which were sufficiently stable to recover and view later with a microscope.

Bonding Mechanism. While the nature of the bonding mechanism which holds hydrophobic particles together in water has not been established with certainty, one of the possible binding forces may be due to the interfacial tension between air and water. If an air bubble is introduced between two parallel flat plates which are made of a hydrophobic material and are submerged in water as shown in Figure 15, the force f due to interfacial tension which pulls the plates towards each other is given by the equation,

$$f = 2 \gamma p \sin \theta \tag{4}$$

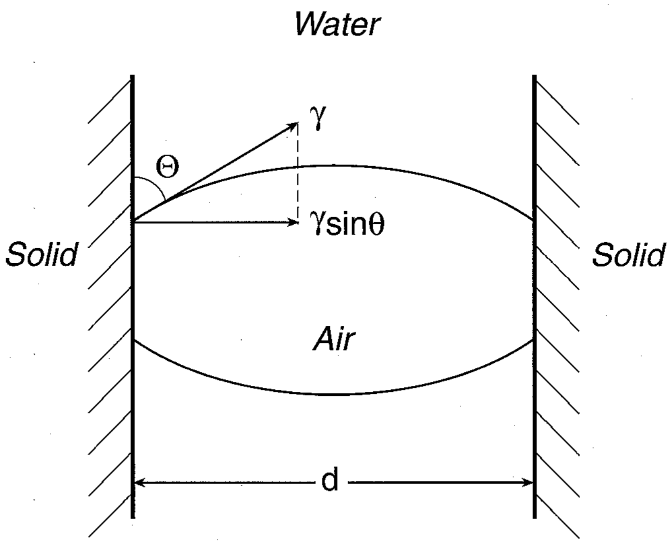


Figure 15. An idealized bridge formed by an air bubble confined between two flat particles immersed in water.

where γ is the air-water interfacial tension, p is the perimeter of the circle of contact of the three phases on the solid surface, and θ is the three-phase contact angle. The same expression has been proposed to account for bonding of two hydrophobic coal particles by an oil bridge (18). Substitution of typical values for γ and θ in equation 4 indicates that the bonding force (f/p) would be similar in magnitude for an air bridge as for an oil bridge.

<u>Discussion of Air Agglomeration</u>. By treating degassed suspensions of various types of hydrophobic particles in water with increasing amounts of air while employing vigorous agitation, it was shown that particle aggregation or agglomeration was promoted by the addition of air. The extent of aggregation was proportional to the amount of air introduced per gram of solids, and it was also proportional to the hydrophobicity of the solids as indicated by the air/water/solid contact angle. In the case of coarse Teflon particles, relatively large agglomerates were produced which may have been held together by air bridges between particles. A model for this type of bonding is presented which is similar to a model proposed

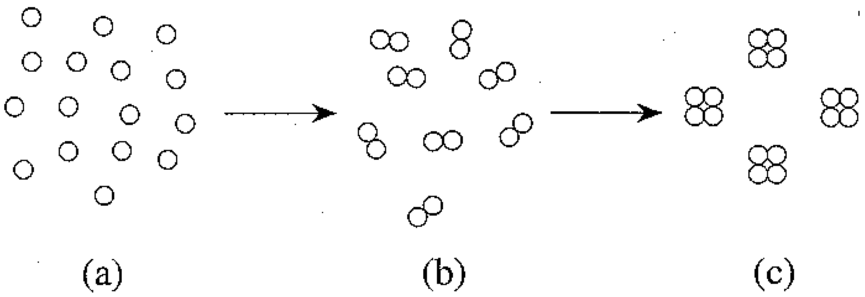


Figure 16. Preliminary stages of a proposed model for representing the formation and growth of agglomerates.

previously for explaining the bonding of oil agglomerated coal particles. In the case of other hydrophobic materials which were small in size compared to the size of gas bubbles, the particles tended to coat the surface of the gas bubbles and to form loose clusters or flocs which were associated with small gas bubbles.

Agglomeration Kinetics of Dilute Suspensions

Technical Basis: For analyzing the kinetics of agglomeration of coal particles, it was assumed that a batch agglomeration test would be conducted starting with a dilute suspension of monosize, spherical particles and that the growth mechanism would involve binary collisions between particles of equal size at each stage of growth as indicated in Figure 16. In other words, it was assumed that all agglomerates would grow at the same rate, and, therefore, at any given instant the suspension would be composed of monosize agglomerates. Furthermore, it was assumed that due to compaction the density of each agglomerate would be essentially the same as that of coal. Consequently, the volume fraction of solids ϕ would remain constant during the process of agglomeration. For a suspension of monosize spherical particles, ϕ would be related to the particle number concentration N and the radius r of an individual particle as follows:

$$\phi = (4/3)\,\pi\, r^3\, N \qquad (5)$$

For an agglomeration process which produces compact, monosize, spherical agglomerates so that ϕ remains constant, the number concentration N of agglomerates of radius r at any instant would be related to the initial concentration N_o of particles of radius r_o as indicated below

$$(4/3)\,\pi\, r^3\, N = (4/3)\,\pi\, r_o^3\, N_o \qquad (6)$$

This equation reduces to:

$$N/N_o = (r_o/r)^3 \qquad (7)$$

For a dilute suspension of monosize spherical particles such that the size of the particles is many times the wavelength of light, the turbidity τ of the suspension is related to particle size and concentration as follows:

$$\tau = 2\,\pi\, r^2\, N \qquad (8)$$

The last two equations are combined to obtain the expression,

$$N/N_o = (\tau/\tau_o)^3 \qquad (9)$$

which relates the particle concentration N and suspension turbidity τ at any time to the initial concentration N_o and turbidity τ_o. Equation 9 is used to determine N from measured values of τ and τ_o and the value of N_o calculated by means of the expression,

$$N_o = 3\, m/4\, \rho\, \pi\, r^3\, V \qquad (10)$$

where m is the total mass of particles, ρ is the particle density, and V is the volume of suspension.

Equation 9 is differentiated with respect to time to provide a means for obtaining the rate of change of particle concentration which is a measure of the rate of agglomeration.

$$\frac{dN}{dt} = \frac{3\,N_o}{\tau_o^3}\,\tau^2\,\frac{d\tau}{dt} \qquad (11)$$

Equations 9 and 11 were modified to account for the fact that only the organic fraction of the coal particles tend to agglomerate. This required assuming that the initial number concentration of organic particles N_o' is related to the initial total number concentration of particles N_o as follows:

$$N_o' = N_o\,(1\text{-}X) \tag{12}$$

where X is the coal ash content. The modified expressions for determining the number concentration of agglomerates N' consisting only of organic particles and the rate of agglomeration of organic particles dN'/dt are shown below.

$$N' = (1 - X)N_o\left[(\tau - X\tau_o)/(1 - X)\tau_o\right]^3 \tag{13}$$

$$\frac{dN'}{dt} = \frac{3\;N_o(\tau - X\tau_o)^2}{(1 - X)^2\,\tau_o^3}\,\frac{d\tau}{dt} \tag{14}$$

Equation 13 was used for estimating the concentration of agglomerates and equation 14 for estimating the rate of agglomeration based on the measured turbidity.

The agglomeration model described above does not differ greatly from the modified Smoluchowski model for orthokinetic flocculation of spherical particles discussed by Gregory (19). According to classical Smoluchowski theory, the rate of flocculation of particles in a uniform shear field is controlled by the rate of collision between particles moving along parallel streamlines. The rate of flocculation was shown to depend on particle size and concentration as indicated below.

$$-\frac{dN}{dt} = \frac{16}{3}\alpha\;G\;r^3\;N^2 \tag{15}$$

In this expression α is the collision efficiency and G is the uniform shear rate. If the volume fraction of particles ϕ remains constant during the process, the preceding equation reduces to,

$$-\frac{dN}{dt} = \frac{4}{\pi}\alpha\;\phi\;G\;N \tag{16}$$

Under such conditions flocculation appears to be a first order rate process with respect to particle concentration. For systems not having a uniform shear field and for turbulent flow conditions, G is replaced by a mean shear rate $\overline{G}$.

Since an actual agglomeration system would not be likely to satisfy all of the assumptions underlying the model described above, the more general and less restrictive rate equation shown below was used to represent the kinetics of agglomeration.

$$-\frac{dN'}{dt} = k(N')^{\beta} \tag{17}$$

In this equation k and β are empirical constants which are likely to depend on coal properties, oil properties and concentration, gas concentration, and the mean shear rate.

To determine k and β and how well equation 17 fit the results of an individual experiment, measured values of τ were first correlated with the corresponding values of t by employing a polynomial equation, usually 10th order. This equation was subsequently differentiated to obtain the rate of change of turbidity for various values of N, and equations 13 and 14 were then used to determine corresponding values of N' and dN'/dt.. These values were used in turn to find k and β by applying linear regression analysis to the data. For this analysis equation 17 was converted to the following form:

$$\ln\left(-\frac{dN'}{dt}\right) = \ln\ k + \beta \ln N' \tag{18}$$

Experimental Method. Coal from the Pittsburgh No. 8 Seam in Belmont County, Ohio, and coal from the Upper Freeport Seam in Indiana County, Pennsylvania, were used for studying the kinetics of agglomeration. The first coal had an ash content of 28% and total sulfur content of 5% while the second coal had an ash content of 14% and total sulfur content of 2%, both on a dry basis. The first coal was much less hydrophobic than the second coal.

After crushing, the coals were wet ground in a stirred ball mill and then stored as a wet paste until used. The weight mean particle size was 14.4 µm for the first coal and 12.7 µm for the second coal.

Agglomeration tests were carried out in a covered cylindrical tank with a flat bottom. The tank had an inside diameter of 15.24 cm (6.0 in.) and height of 15.75 cm (6.2 in.), and it was fitted with four vertical baffles and an agitator. The variable speed agitator was fitted with a 5.08 cm (2.0 in.) diameter, Rushton-type impeller which had six vertical flat blades mounted on a horizontal disk located 2.54 cm (1.0 in.) above the bottom of the tank. An agitator control unit indicated both shaft speed and torque. The measured net volume of the tank fitted with baffles and an agitator was 2870 cm^3 (175 $in.^3$). The turbidity of a particle suspension was determined by continuously pumping a stream of material from the mixing tank through the measuring cell of a photometric dispersion analyzer (PDA 2000 built by Rank Brothers Ltd.) and back to the tank.

To prepare for an agglomeration test, a measured quantity of the previously prepared coal paste was combined with sufficient deionized water to completely fill the mixing tank. Next the agitator and peristaltic pump were operated to eliminate any air pockets. For conducting a run with a known amount of air present, a measured amount of water was withdrawn from the tank while a corresponding amount of air was admitted. The slurry was conditioned to produce a homogeneous state by operating the agitator for 3 min. at the planned operating speed. As agitation was continued, a measured amount of heptane or hexadecane was introduced quickly with a syringe. This marked the beginning of a run. As the run was continued, measurements of the turbidity were made at frequent intervals, and after 20 to 30 min. the run was discontinued.

Experimental Results. A number of batch agglomeration runs were made to study the effects of various parameters on the kinetics of agglomeration of the different types of coal. To demonstrate the importance of having some gas present in the system, several runs were conducted using suspensions of Pittsburgh coal particles which had been degassed first by subjecting the suspensions to a reduced pressure (50-100 mm Hg absolute) for 20 min. The suspensions were then treated with no gas present in the mixing tank. The results of such a run are shown in Figure 17 together with the results of a run made with a suspension of coal which had not been degassed and to which had been added 20 cm^3 of air. It can be seen that the turbidity of the degassed suspension decreased very slowly after heptane was introduced which indicated that the rate of agglomeration was very slow. In contrast the turbidity of the suspension which had not been degassed and which contained added air decreased rapidly indicating rapid agglomeration.

To determine the effects of added air dosage and particle concentration on the actual rate of agglomeration, two series of runs were conducted with suspensions of Pittsburgh coal which had not been degassed. For one series of five runs in which the initial particle concentration varied between 0.1% and 0.5% among runs, 20 cm^3 of air was added to each suspension. For another series of three runs in which the initial particle concentration varied between 0.1% and 0.3% among runs, 50 cm^3 of air was added. For each series the results were fitted by equation 18 to determine the empirical coefficients and the goodness of fit. Figure 18 indicates that in each case most of the data points fell on or close to the straight line determined by linear regression. For 20 cm^3 of added air the line is represented by the equation,

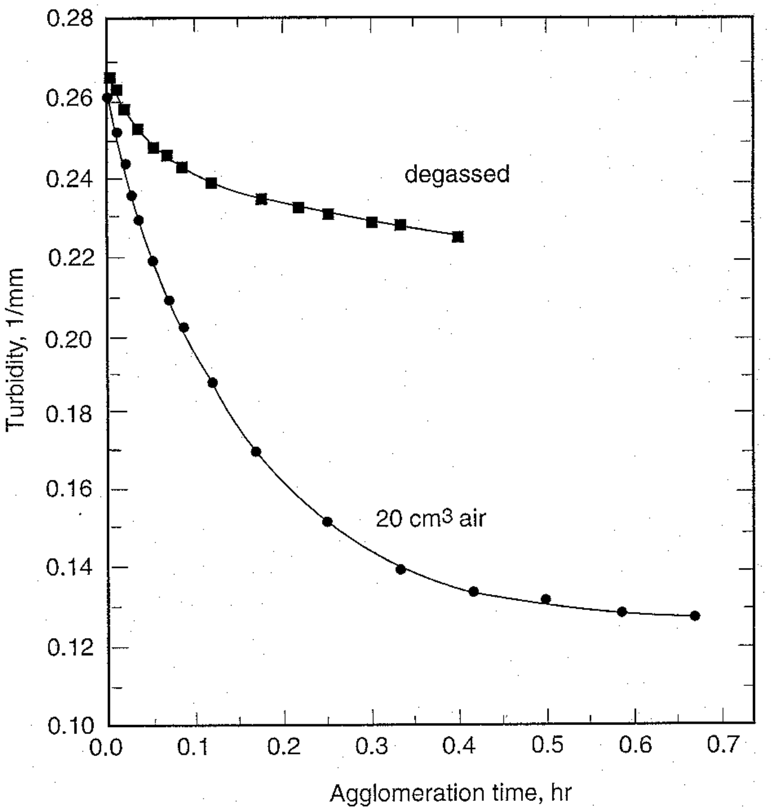


Figure 17. Effect of air on rate of agglomeration of Pittsburgh coal. Conditions: 0.1 w/w% coal, 20 v/w% heptane, 1500 rpm.

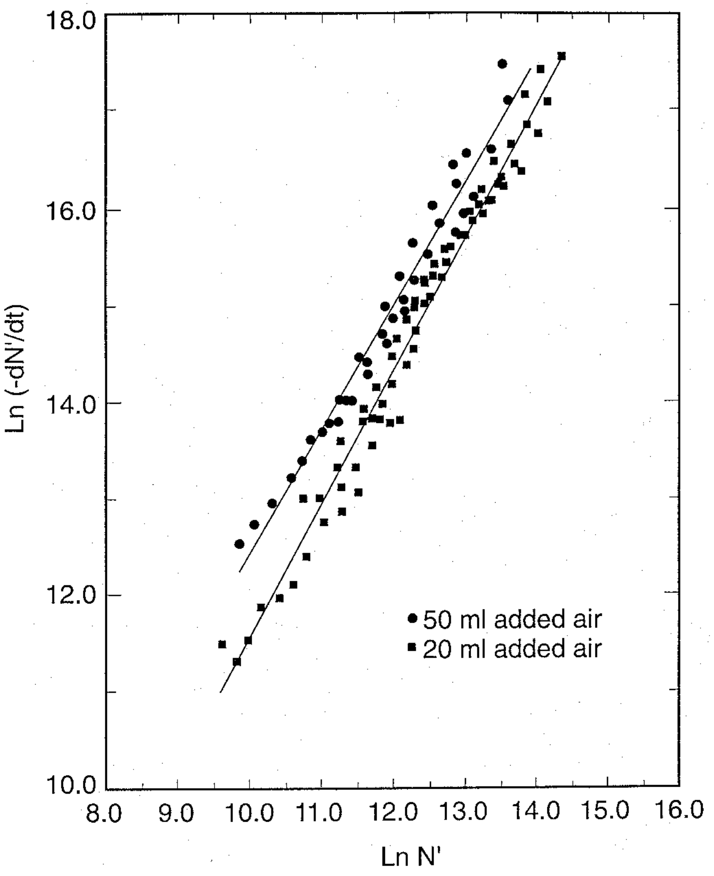


Figure 18. Effects of varying initial particle concentration and added air on rate of agglomeration of Pittsburgh coal. Conditions: 0.1-0.5 w/w% coal, 20 v/w% heptane, 1500 rpm.

$$\ln\left(-\frac{dN'}{dt}\right) = -2.17 + 1.37 \ \ln \ N' \tag{19}$$

and for 50 cm^3 of added air by,

$$\ln\left(-\frac{dN'}{dt}\right) = -0.40 + 1.28 \ \ln \ N' \tag{20}$$

Since the correlation coefficient R was 0.997 in the first case and 0.984 in the second case, an excellent correlation was achieved and the kinetics seemed to be well represented by a rate equation of the form of equation 17. A comparison of the results at the midrange of particle concentration showed that the rate of agglomeration was twice as great with 50 cm^3 of air as with 20 cm^3 of air.

To show the effect of heptane dosage on the rate of agglomeration of Pittsburgh coal, the results of runs made with 10 and 20 v/w% heptane are presented in Figure 19. Again the proposed rate equation fit the results well since a correlation coefficient greater than 0.99 was obtained in each case. The kinetic order was similar for the two cases, whereas the specific rate constant k was 1.69 times greater for the run made with 20 v/w% heptane as for the run made with 10 v/w%. While the rate of agglomeration between runs was affected by heptane concentration, the rate was apparently not affected significantly during a run because of changing heptane concentration. This result together with the apparent independence of the kinetic order on heptane concentration indicates that the rate of agglomeration was not controlled by the rate of collision between oil droplets and coal particles but more likely by the rate of collision between oil-coated particles or between coal aggregates and that the collision efficiency was increased by an increase in heptane dosage.

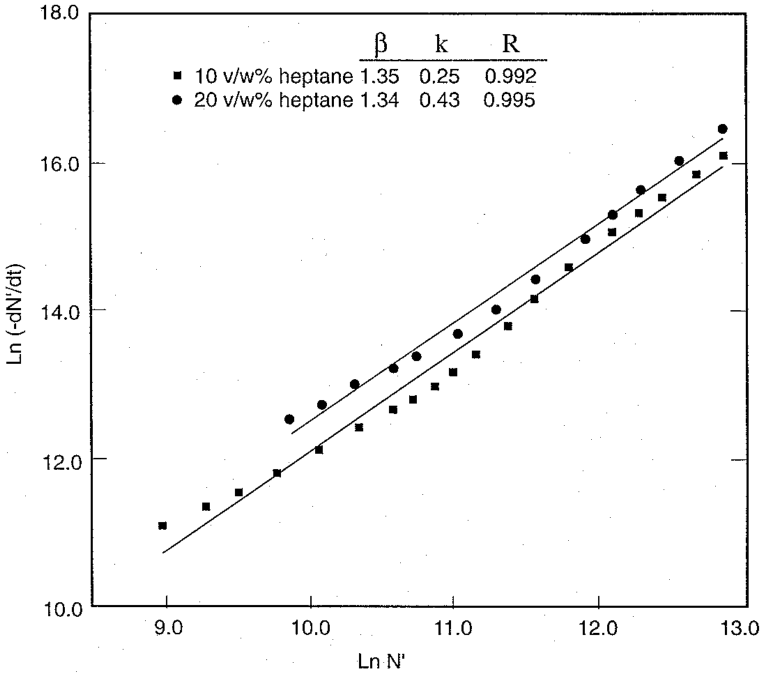


Figure 19. Effect of heptane concentration on rate of agglomeration of Pittsburgh coal. Conditions: 0.1 w/w% coal, 50 cm^3 air, 1500 rpm.

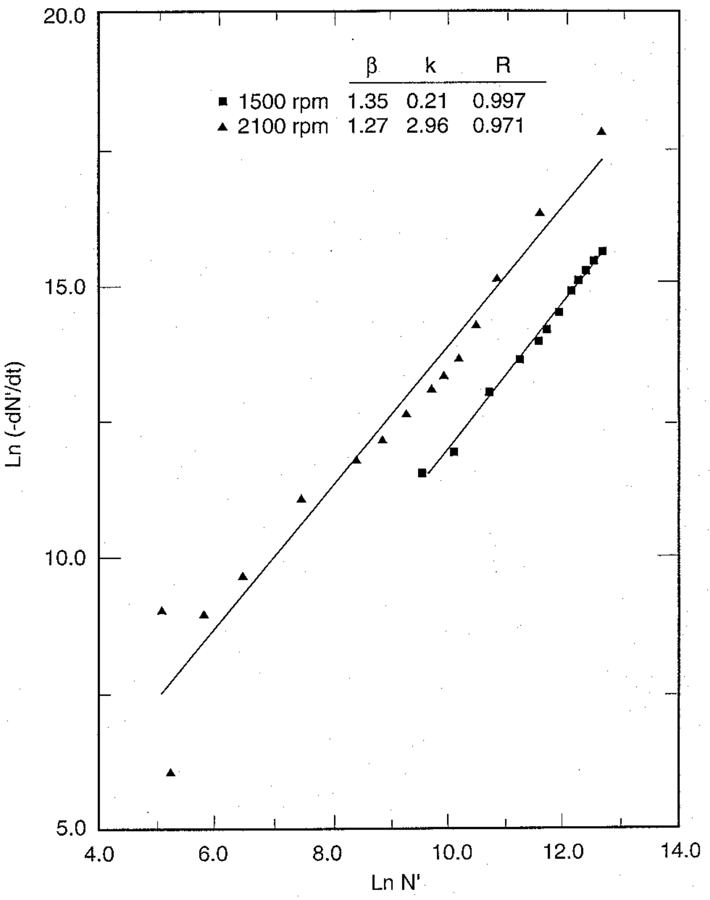


Figure 20. Effect of agitator speed on rate of agglomeration of Pittsburgh coal. Conditions: 0.1 w/w% coal, 20 v/w% heptane, 20 cm^3 air.

To show the effect of agitator speed on the rate of agglomeration of Pittsburgh coal, the results of runs made with different agitator speeds are presented in Figure 20. The results indicate that an increase in agitator speed from 1500 to 2100 rpm produced about a six fold increase in agglomeration rate. Since this is a much larger increase in the rate than the Smoluchowski theory would predict, an increase in shear rate must have had other beneficial effects besides increasing the collision rate between particles.

A number of runs were also made with Upper Freeport coal. The agglomeration of this material was also found to be promoted by a gas as can be seen by the results presented in Figure 21 for two series of runs made with 20 cm^3 of added air and 50 cm^3 of added air, respectively. The first series included eight runs with an initial particle concentration varying between 0.05 and 0.4% among runs, and the second series included three runs with an initial particle concentration varying between 0.1 and 0.24%. Application of linear regression analysis produced the following two equations for the first and second cases, respectively:

For 20 cm^3 air:

$$\ln\left(-\frac{dN'}{dt}\right) = 0.78 + 1.23\ \ln N' \tag{21}$$

For 50 cm^3 of air:

$$\ln\left(-\frac{dN'}{dt}\right) = 1.00 + 1.24\ \ln N' \tag{22}$$

Since the correlation coefficient R was 0.988 in the first case and 0.997 in the second case, the correlation was excellent. A comparison of the results at the midrange of particle concentration showed that the rate of agglomeration was increased approximately 40% by increasing the amount of air from 20 cm^3 to 50 cm^3. While this increase was not as large as

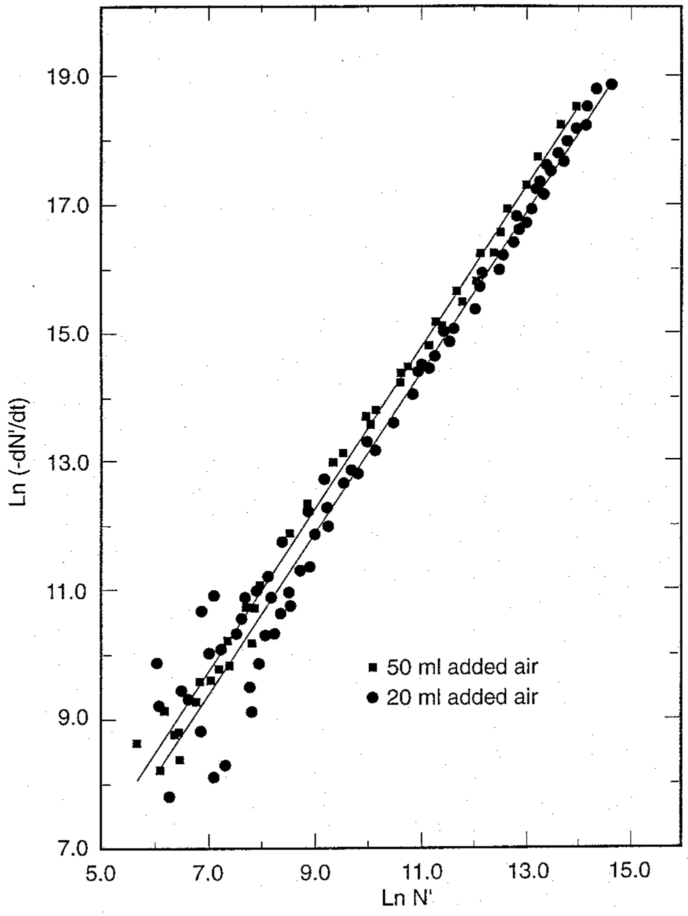


Figure 21. Effects of varying initial particle concentration and added air on rate of agglomeration of Upper Freeport coal. Conditions: 0.05-0.4 w/w% coal, 20 v/w% heptane, 1500 rpm.

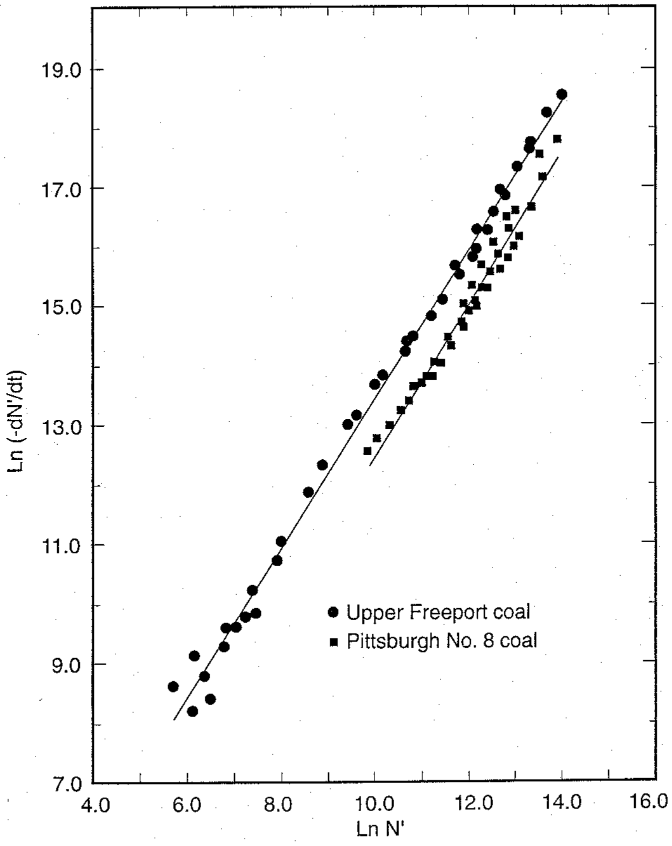


Figure 22. Effect of coal type on rate of agglomeration. Conditions: 0.1 - 0.3 w/w% coal, 20 v/w% heptane, 50 ml air, 1500 rpm.

that observed with the less hydrophobic Pittsburgh coal, it was still significant. It is noteworthy that the kinetic order was virtually the same for the two cases and very similar to that observed for Pittsburgh coal with 50 cm^3 of air (see equation 20).

A comparison of the results achieved with the two types of coal is presented in Figure 22 for the case involving 50 cm^3 of air to promote agglomeration. The lines on this graph correspond to equations 20 and 22 for Pittsburgh coal and Upper Freeport coal, respectively. The rate of agglomeration of Upper Freeport coal was about 2.5 times greater than that of Pittsburgh coal for a particle number concentration corresponding to $\ln N' = 12$. The difference in the rate of agglomeration was due most likely to the difference in the hydrophobicity of the two types of coal.

To see how the rate of agglomeration of Upper Freeport coal was affected by the properties of the agglomerant, the results of runs made with heptane and hexadecane are compared in Figure 23. For the given conditions the rate of agglomeration with hexadecane was approximately half that with heptane at a particle number concentration corresponding to $\ln N' = 12$. This difference may have been due to the larger viscosity of the hexadecane which made it more difficult to disperse. This possibility was supported by another run in which the hexadecane was emulsified first by treating a mixture of the hydrocarbon and water with a high-speed blender before adding it to the coal suspension. The results are shown in Figure 24 together with the results obtained without first emulsifying the hexadecane. As a consequence of emulsification, the rate of agglomeration was increased almost fourfold for $\ln N' = 12$. Since emulsification was carried out in an open blender, it is quite likely that some air was incorporated in the oil in water emulsion which also enhanced the rate of agglomeration.

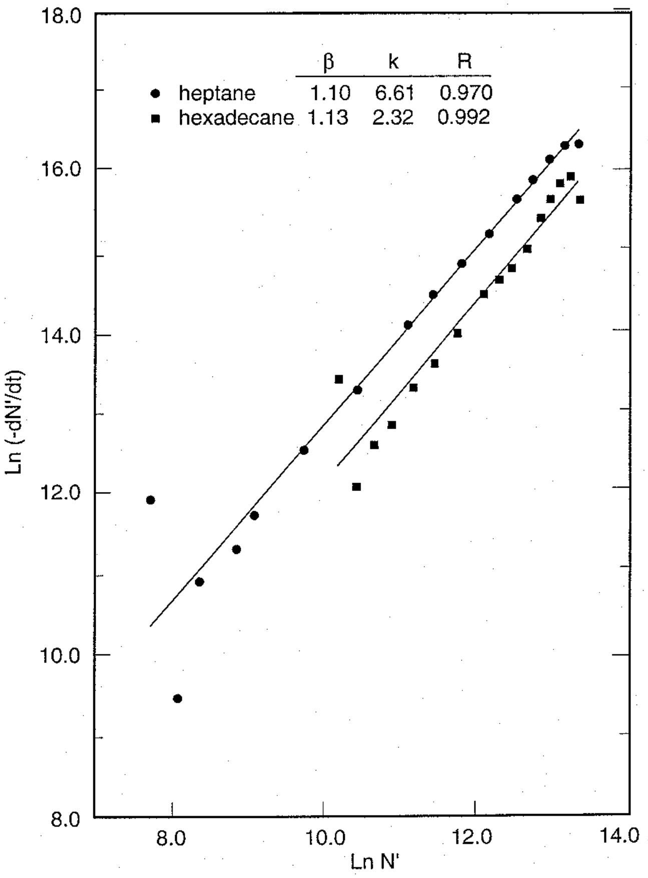


Figure 23. Effect of oil type on rate of agglomeration of Upper Freeport coal. Conditions: 0.1 w/w% coal, 20 v/w% oil, 20 cm^3 air, 1500 rpm.

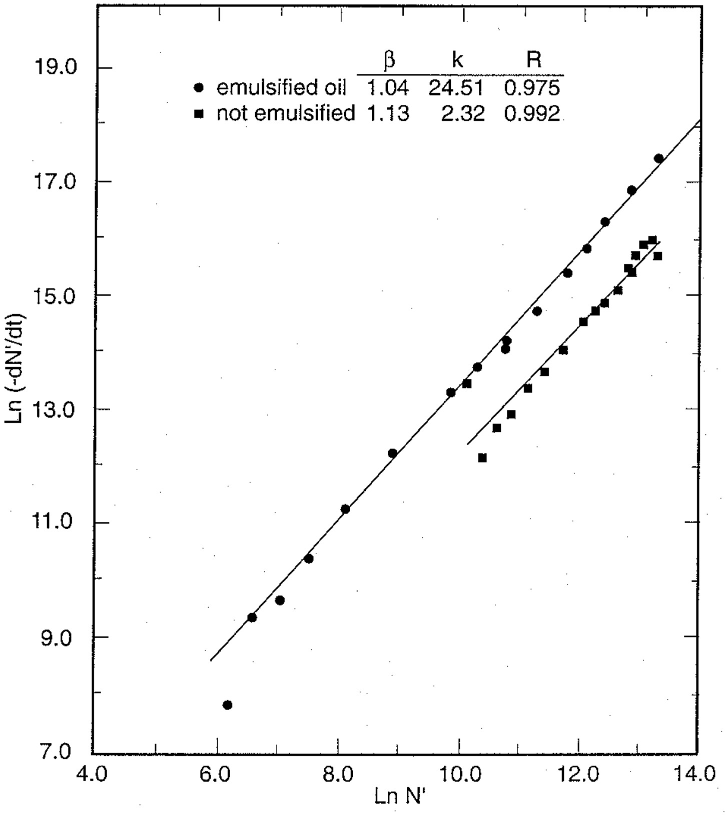


Figure 24. Effect of emulsifying hexadecane before use on rate of agglomeration of Upper Freeport coal. Conditions: 0.1 w/w% coal, 20 v/w% oil, 20 cm^3 air, 1500 rpm.

Discussion of Kinetics. A semiempirical rate equation used for correlating and analyzing the results of batch agglomeration tests seemed to provide an adequate representation of the kinetic data collected during most individual runs or a series of runs made with different initial particle concentrations. In most cases the kinetic order with respect to particle concentration was between 1.0 and 1.3 which was fairly consistent with the assumptions underlying the proposed mechanism and measurement technique. The rate of agglomeration seemed to be controlled by the rate of collision between oil-coated particles or between coal aggregates rather than between oil droplets and coal particles. The rate of agglomeration increased as more air was added to the system or as the agglomerant concentration or agitator speed was increased. The rate was also greater for the more hydrophobic Upper Freeport coal than for the less hydrophobic Pittsburgh No. 8 coal. In addition, a greater rate was achieved with heptane than with hexadecane. The rate achieved with hexadecane was enhanced considerably by first emulsifying the material in water which also probably introduced more gas micro-bubbles into the system.

RESULTS ACHIEVED WITH CONCENTRATED SUSPENSIONS

Several preliminary experiments showed that the agglomeration of concentrated coal suspensions was accompanied by changes in agitator torque which could be used to monitor the progress of agglomeration during a batch agglomeration test conducted at a constant agitator speed. For a wide range of experimental conditions this method provided a convenient way to measure the minimum time t_e required to produce compact, spherical agglomerates. For some conditions the method had to be supplemented by direct observation which involved removing small samples from the system at frequent intervals and examining them with a microscope. In addition to determining t_e, agglomeration tests were conducted to determine the size of the agglomerates produced as well as coal recovery and ash rejection.

Several series of agglomeration tests were conducted to determine the effects of a number of different parameters. The results were analyzed and correlated by employing empirical relations, some of which were based on dimensional analysis and engineering similitude.

Agglomeration Tests, Preliminary Series A

Coal from the Pittsburgh No. 8 Seam was used for the first preliminary series of agglomeration tests or runs. There were 35 runs in this series. A large batch of coal was reduced with a double-roll crusher to approximately 2 mm size particles and then divided into smaller portions by riffling. These portions were stored under argon until needed. Sometime before an agglomeration test or a group of tests, one or more of the smaller portions was recrushed with the double-roll crusher to pass a 1.4 mm screen. The material was then ground in a stirred ball mill to produce particles in the 1 to 10 µm size range. The slurry from the ball mill was partially dewatered to produce a paste which was stored in a refrigerator set at 5°C until used.

For each agglomeration test the coal was suspended in deionized water and the suspension was degassed for 20 min. by applying a vacuum corresponding to –95 kPa. The test was carried out as described previously using pure i-octane from Burdick and Jackson Laboratories as an agglomerant. The suspension of coal, oil and water was stirred for 5 min. before a specific amount of air was introduced. Conditioning without air was conducted by operating the agitator at the speed required for the run except for runs made with agitator speeds greater than 1800 rpm. In those cases, the speed was limited to 1800 rpm for conditioning.

The specific combination of conditions used for each test in the series is shown in Table 5. It can be seen that three sizes of mixing tanks were employed as well as three sizes of impellers. Various agitator speeds ranging from 1100 to 2450 rpm were also used.

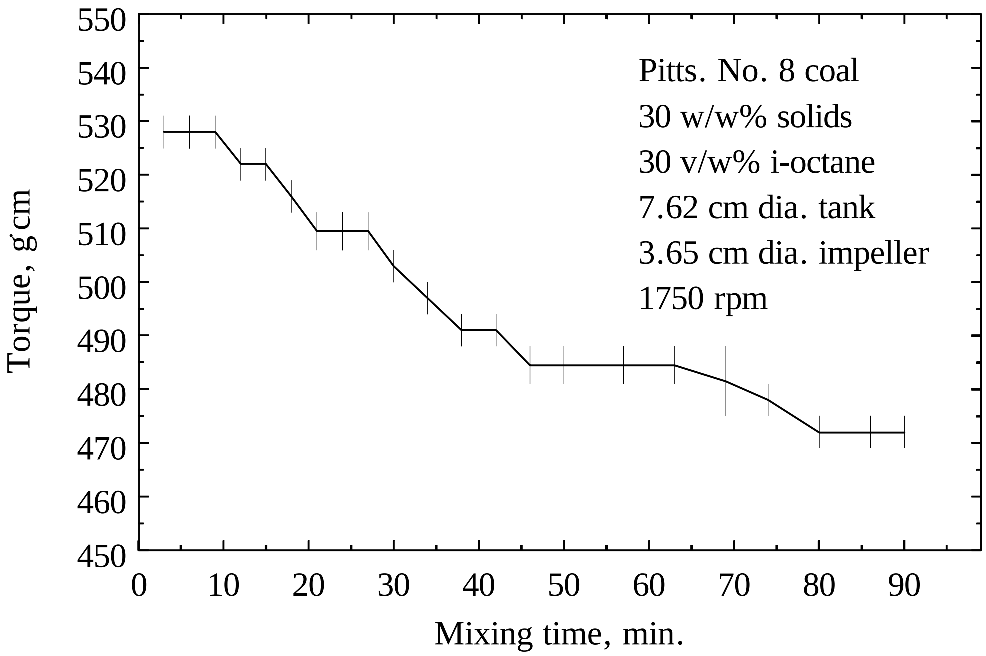
Table 5. Experimental conditions and results for batch agglomeration tests with Pittsburgh No. 8 coal and i-octane: Preliminary Series A

Run No.	T, cm	D, cm	N, rpm	S, m/s	Solids, w/w%	Oil, v/w%	Air, v/w%	t_e min.	t_t min	P/V, kw/m^3	Aggl. Size, mm
1	15.24	6.35	1300	4.32	30	20	9.0	32	105	17.00	0.09-0.12
2	15.24	5.08	2400	6.38	30	20	9.0	15	105	27.30	0.08-0.11
3	15.24	5.08	2000	5.32	30	20	9.0	23	105	15.80	0.06-0.08
4	15.24	5.08	1600	4.26	30	20	9.0	70	105	7.77	0.06-0.08
5	15.24	5.08	1300	3.46	30	20	9.0	87	115	3.96	0.05-0.10
6	15.24	5.08	1100	2.93	30	20	9.0	540	570	2.58	0.04-0.08
7	11.43	3.65	1600	3.06	30	20	9.0	83	115	4.29	0.04-0.08
8	7.62	5.08	1600	4.26	30	20	9.0	28	30	74.16	0.10-0.15
9	7.62	3.65	1600	3.06	30	20	9.0	80	95	17.22	0.04-0.08
10	15.24	3.65	2400	4.59	30	20	9.0	35	105	5.98	0.05-0.07
11	15.24	3.65	2000	4.44	30	20	9.0	89	120	3.22	0.05-0.10
12	15.24	3.65	1600	3.06	30	20	9.0	315	335	1.87	0.06-0.09
13	7.62	3.65	1750	3.34	30	20	9.0	33	95	21.54	0.10-0.20
14	11.43	5.08	1080	2.87	30	20	9.0	125	150	5.52	- - -
15	7.62	3.65	1675	3.20	30	20	9.0	40	95	18.03	0.08-0.15
16	7.62	5.08	1080	2.87	30	20	9.0	>105	105	21.20	0.08-0.20
17	7.62	5.08	1150	3.06	30	20	9.0	120	120	24.93	0.04-0.10
18	7.62	5.08	1300	3.46	30	20	9.0	63	90	37.08	0.06-0.10
19	7.62	3.65	2000	3.82	30	20	9.0	- - -	11	- - -	- - -
20	11.43	5.08	2450	6.52	30	20	9.0	- - -	13	- - -	- - -
21	7.62	3.65	1750	3.34	30	30	9.0	20	45	21.69	0.20-0.40
22	7.62	3.65	1550	2.96	30	30	9.0	30	45	14.85	0.20-0.40
23	7.62	3.65	2200	4.20	30	30	9.0	4.5	11	39.79	0.15-0.30
24	11.43	5.08	2000	5.32	30	30	9.0	6.5	25	42.43	0.25-0.40
25	11.43	3.65	1850	3.54	30	30	9.0	39	65	7.02	0.15-0.30
26	7.62	3.65	1750	3.34	30	20	18.0	22	50	19.30	0.08-0.10
27	7.62	3.65	1750	3.34	30	20	4.5	100	120	19.82	0.05-0.08
28	7.62	3.65	2200	4.20	30	20	4.5	22	45	38.49	0.05-0.08
29	7.62	5.08	1400	3.72	30	20	4.5	95	95	47.87	0.05-0.08
30	11.43	5.08	2000	5.32	30	20	4.5	17	25	39.62	0.04-0.08
31	11.43	3.65	2400	4.59	30	20	4.5	25	95	14.15	0.05-0.09
32	7.62	3.65	2200	4.20	30	20	2.3	23	38	38.49	0.04-0.08
33	7.62	3.65	1750	3.34	30	20	2.3	151	160	19.82	0.05-0.08
34	7.62	3.65	2200	4.20	30	20	18.0	5	15	37.38	0.06-0.10
35	7.62	3.65	1750	3.34	30	30	0	- - -	85	- - -	- - -

Different amounts of air ranging from 0 to 18 v/w% and i-octane concentrations of either 20 v/w% or 30 v/w% were utilized. Both the air and oil concentrations were based on the weight of solids supplied to the mixer. Therefore, an oil concentration of 20 v/w% corresponded to 20 ml oil/100 g solids. The solids concentration employed in each test was 30 w/w% based on the weight of water.

A preliminary analysis of the results revealed a great deal about the nature and basic characteristics of the agglomeration process. Air had to be present in the mixing system for agglomeration to take place. This can be seen from the results of run 35 conducted without air. Figure 25 indicates that the agitator torque decreased more or less steadily throughout the run. Previous results had shown that particle agglomeration was always accompanied by a marked increase in agitator torque. Furthermore, in run 35 when the suspension of coal particles and i-octane was examined after 90 min. of agitation, only free particles and microflocs were observed. In sharp contrast to these results were the results of runs made with air present in the system which included all other runs. In every run where air was present, there was evidence of agglomeration. Even in run 32 where the concentration of air was only 2.25 v/w%, agglomeration was indicated by a sharp increase in agitator torque which occurred 20 min. after air was introduced (see Figure 26). Agglomeration was confirmed by the presence of agglomerates in a sample taken at the time when the torque was a maximum. The agglomerates ranged from 0.04 to 0.08 mm in diameter.

A curve representing changes in agitator torque as a function of time for a batch agglomeration test usually included several maxima and minima, and it indicated that the



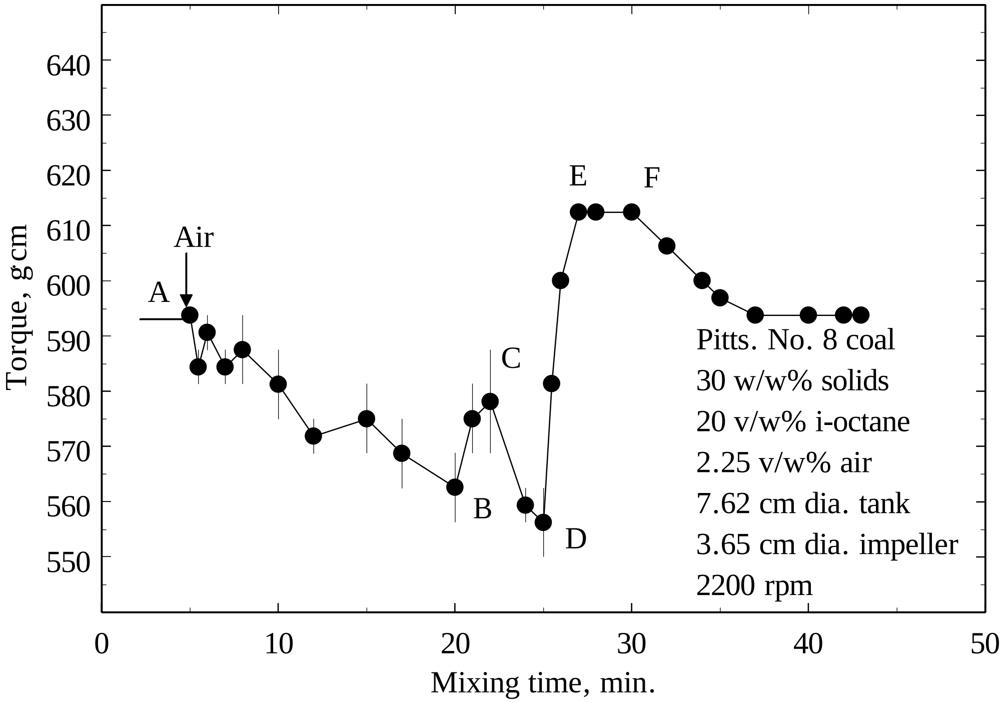


Figure 25. Changes in agitator torque in run 35 made without air.

Figure 26. Changes in agitator torque in run 32 made with 2.25% v/w% air.

process was relatively complex and consisted of several stages. Examples of such curves are presented in Figures 26 and 27. By examining samples of suspensions taken at various stages of agglomeration it was possible to relate the changes in agitator torque to physical changes in the suspensions. For the purpose of discussion, key turning points in agitator torque have been identified by various letters in Figures 26 and 27. The initial conditioning period when oil was dispersed is identified by letter A. During this period and depending on the intensity of agitation, oil droplets usually became covered and stabilized by hydrophobic coal particles and some microflocs were produced. However, the majority of the particles generally remained freely suspended. After air was introduced, there was a drop in agitator torque as the air was dispersed into small bubbles which apparently reduced the viscosity of the suspension, and the torque decreased to a minimum value at point B. As the process continued from point B to point C, aggregates which contained oil and hydrophobic coal particles were drawn to and accumulated on the surface of bubbles because the bubbles were highly hydrophobic. The accumulation of particles at the air/water interface produced larger flocs and flakes which may have become detached from the bubbles after reaching a certain size. This process ultimately reduced the concentration of free coal particles and microflocs to a level where there was a marked change in the color of the suspension from black to grey. The color change became very noticeable at point C. Previous investigators have referred to the color change as the “inversion point.”

Between point C and point D the agitator torque dropped which may have been the result of several factors. One possible factor was the release of gas bubbles by the detachment of flocs and flakes. Another possible factor was the formation of a gel-like structure which took place between C and D. The structure had the appearance of flocks interconnected by gas bubbles. This structure tended to limit circulation of the suspension to a region near the impeller. At point

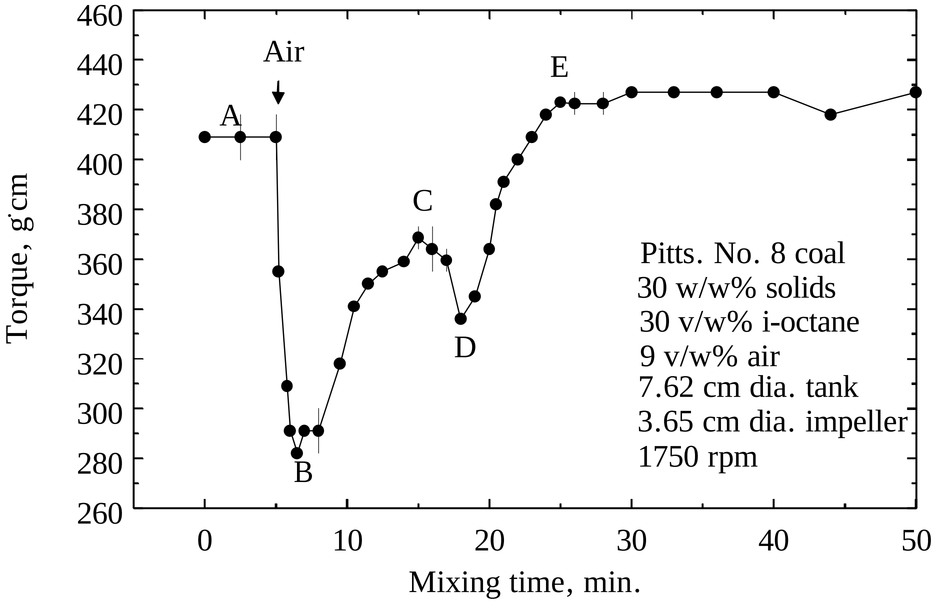


Figure 27. Changes in agitator torque in run 21 made with 9 v/w% air.

D most of the hydrophobic particles appeared to be incorporated into relatively large flocs or loose agglomerates which were converted subsequently into spherical agglomerates between D and E.

Subjecting the agglomerates to further agitation beyond point E increased the sphericity of individual agglomerates and produced a narrower size distribution of agglomerates. Also the agglomerates seemed to become more hydrophobic which could have been the result of oil being forced to the surface of the agglomerates. Consequently, under some conditions the agglomerates combined to form larger agglomerates with a framboidal structure. The consolidation of individual agglomerates had little effect on agitator torque as can be seen by the results indicated in Figure 27, which show that the torque remained almost constant following point E.

While most of the runs in the series produced a pattern of torque changes similar to those indicated by Figures 26 and 27, there was considerable variation among runs in the time required to complete the various steps in the agglomeration process and in the torque measured at the key turning points in the process. The time required to reach point E was especially important since it seemed to represent the minimum time required to produce compact, spherical agglomerates. For the preliminary series of runs, the time interval between the point when air was introduced and point E was taken to be the minimum agglomeration time t_e.

Values of t_e are presented in Table 5 for the different agglomeration tests together with the total time t_t each suspension was stirred after air was introduced. The size range of the agglomerates observed at the end of each test is also indicated. For a given oil concentration the variation in the size range of the agglomerates was relatively small among different runs. However, the agglomerates produced with 30 v/w% oil were noticeably larger than those produced with 20 v/w% oil.

The agitator power input per unit volume (P/V) was a key parameter since it seemed to affect the rate of agglomeration and would have an important effect on the operating cost of any industrial application of the process. Values of P/V based on measured agitator torque and speed at point E are indicated in Table 5. The operating conditions at point E seem especially relevant since a continuous industrial process would probably be carried out under conditions such that most of the coal particles in the system would be largely in the form of compact, spherical agglomerates as also seemed to be the case for the coal present in the laboratory batch agglomerator at point E.

It is widely accepted that the power requirement P for an agitator fitted with an impeller having a diameter D and operating in the turbulent flow regime can be predicted by the following relation (20):

$$P = N_p \, \rho \, N^3 D^5 / g_c \tag{23}$$

where N_p is the dimensionless power number which is constant for a given agitator operating under turbulent flow conditions, ρ is the density of the fluid, N is the rotational speed of the impeller, and g_c is a dimensional constant. The volume V of a cylindrical mixing tank constructed so that its height is equal to its diameter is given by the expression,

$$V = (\pi / 4)T^3 \tag{24}$$

where T is the inside tank diameter. Dividing equation 23 by equation 24 leads to the following relation:

$$P/V = \left(4\rho N_p / \pi g_c\right) N^3 D^5 / T^3 \tag{25}$$

Consequently, for a fixed impeller design and constant fluid density, the preceding expression reduces to the following:

$$P/V = KN^3 D^5 / T^3 \tag{26}$$

$$\text{where } K = 4\,\rho N_p / \pi g_c$$

To see whether the observed values of P/V reported in Table 5 were related to the system properties as suggested by equation 26, the following expression was fitted to the data by linear regression:

$$\ln(P/V) = \alpha + \beta \ln\left(N^3 D^5 / T^3\right) \tag{27}$$

This produced the equation,

$$P/V = 4.99\,\left(N^3 D^5 / T^3\right)^{1.02} \tag{28}$$

The fit was excellent since the correlation coefficient R was 0.993 or the coefficient of determination R^2 was 0.987. Furthermore, the 95% confidence interval for β was 1.02 ± 0.04 which encompassed the generally accepted value of 1.00. Therefore, there was good agreement between the form of equation 28 and the form of equation 23.

Based on the value K = 4.99 in equation 28 and an estimated slurry density of 1.06 g/cm^3 for a slurry containing 30 w/v% solids, 20 v/w% i-octane, and 9 v/w% air, the value of the power number N_p was found to be 3.70. This value is smaller than the value of 5 reported for standard or conventional systems involving the same type of turbine impeller operating in a cylindrical tank with four baffles (20). However, the present system differed from a standard or conventional system in several ways. In the present system the single impeller was located midway between the top and bottom of the tank, while more conventional practice is to locate the impeller at a height above the tank bottom equal to the impeller diameter. In addition, the upper surface of the liquid suspension was constrained by the top of the tank in the present case, whereas in more conventional systems there is usually a gas space between the surface of the liquid and the top of the tank.

Various ways of correlating and predicting agglomeration system performance were considered and tested. For the preliminary series of agglomeration tests the best correlation was obtained by fitting the following empirical equation to the experimental data:

$$t_e = aX_1^b X_2^c (D/T)^d (P/V)^e \tag{29}$$

where X_1 and X_2 are the concentrations of oil and air, respectively, in ml/g coal, D/T is dimensionless, P/V is in kW/m^3, and t_e is in minutes. This equation was converted to a linear form by taking the logarithm of both sides, and the resulting expression was used for actual data fitting by application of multiple linear regression which produced the following result:

$$t_e = 83.46 \ X_1^{-1.186} X_2^{-0.537} (D/T)^{0.950} (P/V)^{-1.217} \tag{30}$$

The fit was good since the correlation coefficient R was 0.939 or the coefficient of determination R^2 was 0.882. In addition, each of the independent terms on the right hand side of the equation seemed to make a statistically significant contribution to t_e. However, to achieve a fit this good it was necessary to disregard several outlying points and utilize only 24 of the observations listed in Table 5.

Dimensional analysis was considered as a means for developing a suitable correlation for t_e and this led to the following expression:

$$Nt_e = f\left(X_1, X_2, D/T, ND^2\,\rho/\mu\right) \qquad (31)$$

Utilizing this expression presents a problem because it requires that the viscosity μ of the suspension undergoing agglomeration be known. Unfortunately, the rheological properties of the suspension are extremely complex since there are at least five phases present: coal particles, mineral particles, water, oil, and gas. In addition, the rheological properties change as coal particles are agglomerated, and this change is reflected by the agitator torque which varies during agglomeration.

For a preliminary assessment of the value of dimensional analysis, it was assumed that both ρ and μ are constant. Utilizing the same 24 observations as before and applying multiple linear regression produced the following equation:

$$Nt_e = 9.67\ X_1^{-1.835}\,X_2^{-0.304}\left(D/T\right)^{-1.897}\left(N^* D^2\right)^{-1.12} \qquad (32)$$

The assumed constants ρ and μ are embedded in the first constant. In equation 32, N^* is the agitator speed in revolutions per second, whereas N is in revolutions per minute as it is in other equations. Also D is in meters. Equation 32 did not prove as suitable as equation 30 for representing the results of the series of agglomeration tests because it had a correlation coefficient of 0.743 which was appreciably lower than that for equation 30. Furthermore, X_2 and possibly X_1 did not made a statistically significant contribution to t_e. Therefore, the assumption of constant viscosity does not appear valid.

Agglomeration Tests, Preliminary Series B

Coal from the Pittsburgh No. 8 Seam was also used for the second preliminary series of agglomeration tests. The coal was prepared and stored as in the previous series. Measurement of the particle size distribution by automated image analysis showed that the wet milled coal had a projected area mean particle diameter of 4.82 μm with a particle diameter range of 1 to 31 μm.

The agglomeration tests were conducted as before except that the agglomerated product and tailings were recovered and analyzed. For each agglomeration test the aqueous suspension of coal particles was first degassed by applying a vacuum, and then it was dosed with pure i-octane from the Burdick and Jackson Laboratories. The mixing tank was subsequently filled with water, and the suspension was conditioned for 5 min. by running the agitator at the specified test speed. A known volume of air was admitted by withdrawing a corresponding volume of slurry with a syringe. This started the process of agglomeration, which was continued by operating the agitator at constant speed and by cooling the mixing tank to maintain a constant temperature. During the run the agitator torque was recorded. At the end of a run a sample of the suspension was collected and later analyzed with a microscope. The remaining suspension was then diluted with an equal volume of water and separated with a 250 μm screen. The agglomerated product and the tailings were recovered separately and then were dried and weighed. Later the ash content of the product and tailings was determined.

The conditions employed and the results obtained in this series of agglomeration tests or runs are presented in Table 6. It can be seen that the following parameters were varied among the tests: tank size, impeller diameter and speed, solids concentration, oil concentration, air concentration, and total treatment time. The ratio of tank diameter to impeller diameter was nearly the same for the two mixing tanks employed since the ratio was equal to 2.1 for the smaller tank and 2.25 for the larger tank. Most of the runs were conducted with a solids concentration of 30 w/w%, while only two runs were conducted with a solids concentration of 20 w/w%.

The results of this series of tests confirmed certain aspects of the previous series. Thus it was observed that agitator torque varied during the various stages of agglomeration in much the same way that was indicated by Figures 26 and 27 above. The similarity can be seen by

Table 6. Experimental conditions and results of batch agglomeration tests with Pittsburgh No. 8 coal and i-octane: Preliminary Series B

Run, No.	T, cm	D, cm	N, rpm	S, m/s	Solids, w/w%	Oil, v/w%	Air, v/w%	t_t min.	Aggl. Size, mm	Ash, [a] w/w%	Ash Rej.,[b] %	Recov.[c] %
17	7.62	3.65	1600	3.05	30	30	9.0	35	0.2	11.87	64.60	95.40
18	7.62	3.65	1600	3.05	30	30	9.0	45	0.2	11.68	66.50	94.20
20	7.62	3.65	1600	3.05	30	30	9.0	80	2.5	8.19	77.40	96.80
22	7.62	3.65	1600	3.05	30	30	9.0	80	2.5	7.82	79.30	96.10
24	7.62	3.65	2000	3.82	30	30	9.0	65	2.0~3.5	7.95	78.40	95.50
25	7.62	3.65	2000	3.82	30	30	9.0	45	0.2-0.3	10.28	71.40	92.20
26	7.62	3.65	2000	3.82	30	30	9.0	25	0.15~0.2	11.87	67.00	92.90
27	7.62	3.65	2000	3.82	30	30	9.0	10	big flocs	15.90	55.30	88.80
28	7.62	3.65	2000	3.82	30	20	9.0	130	0.15~0.22	11.20	68.70	94.20
29	7.62	3.65	2200	4.20	30	20	9.0	132	0.08~0.1	12.98	64.90	90.40
30	7.62	3.65	1800	3.44	30	20	9.0	130	flocs	19.89	65.50	52.90
32	7.62	3.65	1600	3.05	30	20	18.0	85	0.05~0.1	12.81	70.60	76.60
33	7.62	3.65	1800	3.44	30	20	18.0	85	0.05~0.1	16.68	56.80	85.70
34	7.62	3.65	2000	3.82	30	20	18.0	85	flocs	16.72	51.80	91.60
35	7.62	3.65	2200	4.20	30	20	18.0	25	flocs	15.91	60.90	78.40
36	7.62	3.65	2200	4.20	30	20	18.0	85	0.08~0.1	12.42	65.40	94.30
39	7.62	3.65	2000	3.82	30	20	18.0	55	flocs	15.34	67.50	64.70
40	7.62	3.65	2000	3.82	30	20	18.0	70	0.05~0.08	15.08	61.30	85.70
41	7.62	3.65	2000	3.82	30	20	18.0	15	flocs	17.89	73.70	45.60
42	7.62	3.65	2000	3.82	30	30	18.0	85	0.25~0.8	9.30	73.30	96.40
43	7.62	3.65	2000	3.82	30	30	18.0	45	0.2~0.3	9.50	72.80	96.80
44	7.62	3.65	2000	3.82	30	30	4.5	115	2	7.72	79.30	95.30
45	7.62	3.65	2000	3.82	30	30	4.5	85	1.8~2.0	7.92	78.50	96.00
47	7.62	3.65	2000	3.82	30	30	4.5	45	0.2~0.25	12.90	62.00	88.20
48	7.62	3.65	2000	3.82	30	20	4.5	85	0.05~0.1	21.24	36.90	71.50
49	11.43	5.08	1600	4.26	30	30	10.0	40	0.20-0.30	10.36	68.87	97.36
50	11.43	5.08	1600	4.26	30	30	9.0	45	0.20-0.25	10.52	70.22	95.53
51	11.43	5.08	1600	4.26	30	30	9.0	85	0.40-0.60	8.64	74.27	96.51
52	11.43	5.08	1600	4.26	30	30	9.0	10	0.15-0.20	8.95	69.43	94.34
53	11.43	5.08	1600	4.26	30	30	9.0	25	0.18-0.22	13.22	64.21	95.39
54	11.43	5.08	1600	4.26	30	30	15.0	25	0.20-0.22	8.90	73.96	95.52
55	11.43	5.08	1750	4.65	30	30	9.0	85	0.40-0.60	8.79	74.50	96.90

Table 6. Continued

Run, No.	T, cm	D, cm	N, rpm	S, m/s	Solids, w/w%	Oil, v/w%	Air, v/w%	t_t min.	Aggl. Size, mm	Ash, [a] w/w%	Ash Rej.,[b] %	Recov.[c] %
56	11.43	5.08	1750	4.65	30	30	9.0	25	0.15-0.20	8.21	74.89	95.06
57	11.43	5.08	1750	4.65	30	30	9.0	45	0.25-0.32	8.45	74.04	97.14
58	11.43	5.08	1750	4.65	30	30	9.0	10	0.12-0.15	11.04	65.94	93.36
59	11.43	5.08	1450	3.86	30	30	9.0	25	0.10-0.15	13.01	61.30	90.93
60	11.43	5.08	1450	3.86	30	30	9.0	10	0.08-0.10	18.97	45.97	87.49
61	11.43	5.08	1450	3.86	30	30	9.0	85	1.20-4.00	8.33	77.05	96.72
62	11.43	5.08	1450	3.86	30	30	9.0	45	0.15-0.25	10.84	68.87	92.25
63	11.43	5.08	1450	3.86	30	30	4.5	85	0.25-0.40	9.78	72.77	96.44
64	11.43	5.08	1600	4.26	30	20	9.0	85	0.09-0.12	14.98	62.32	83.18
65	11.43	5.08	1600	4.26	30	20	9.0	55	0.07-0.10	15.35	64.11	75.36
66	11.43	5.08	1750	4.65	30	20	9.0	85	0.08-0.12	14.59	60.37	87.47
67	11.43	5.08	1750	4.65	30	20	9.0	55	0.07-0.10	15.51	60.65	80.92
68	11.43	5.08	1750	4.65	30	20	9.0	25	0.05-0.07	19.59	58.17	65.63
69	11.43	5.08	1450	3.86	30	20	9.0	85	0.07-0.12	16.45	58.90	83.34
70	11.43	5.08	1450	3.86	30	20	9.0	55	0.06-0.10	17.51	59.70	74.97
71	11.43	5.08	1450	3.86	30	20	9.0	25	0.05-0.06	22.68	38.57	81.77
72	11.43	5.08	1600	4.26	30	20	4.5	85	0.05-0.10	17.91	53.45	81.64
73	11.43	5.08	1600	4.26	30	20	4.5	55	0.03-0.09	18.81	55.08	75.04
74	11.43	5.08	1750	4.65	30	20	4.5	55	0.05-0.10	18.16	57.41	74.22
75	11.43	5.08	1450	3.86	30	20	4.5	55	(flocs)	22.48	93.02	9.21
77	11.43	5.08	1600	4.26	30	20	18.0	55	0.08-0.12	13.82	62.94	88.15
78	11.43	5.08	1450	3.86	30	20	18.0	85	0.08-0.11	14.05	62.93	85.00
79	11.43	5.08	1750	4.65	30	20	18.0	85	0.07-0.12	12.54	64.67	91.98
80	11.43	5.08	1600	4.26	30	20	18.0	85	0.07-0.11	12.97	64.76	91.91
81	11.43	5.08	1450	3.86	30	20	9.0	25	0.04-0.06	20.50	43.90	82.39
82	11.43	5.08	1750	4.65	20	30	9.0	85	0.25-0.30	8.37	76.50	96.63
83	11.43	5.08	1600	4.26	20	30	9.0	85	0.30	8.42	77.64	96.63

[a] Ash content of agglomerates

[b] Ash rejected to tailings

[c] Coal recovery on a dry-ash-free basis

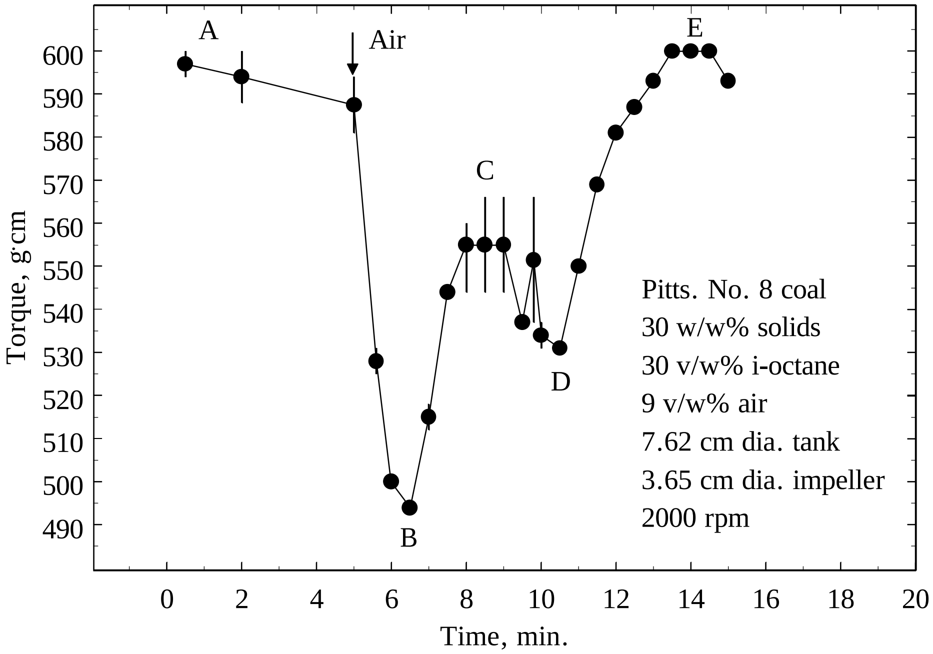


Figure 28. Changes in agitator torque during run 27.

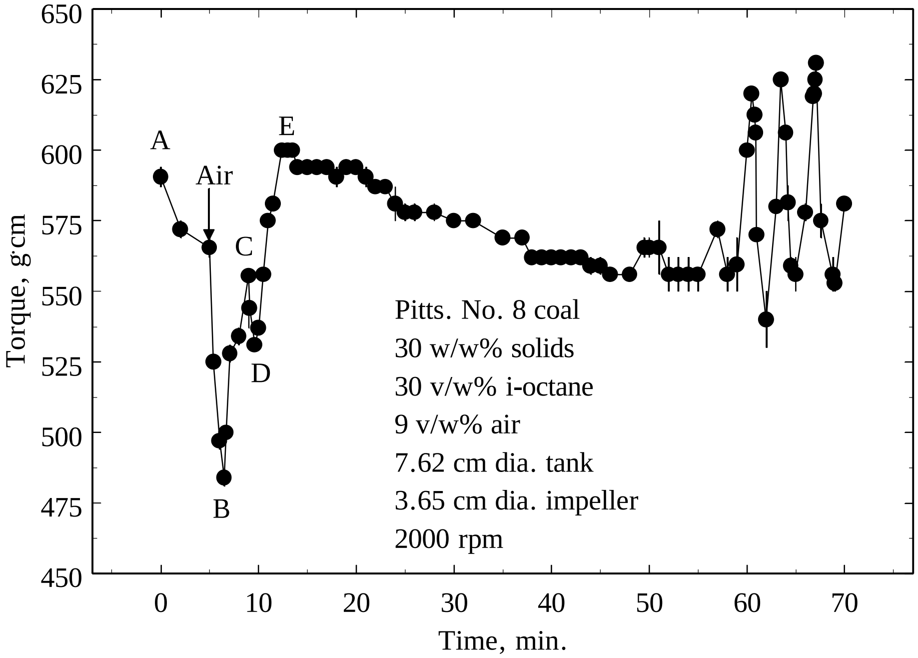


Figure 29. Changes in agitator torque during run 24.

comparing the present results presented in Figures 28 and 29 with the previous results. As before, after the introduction of air there was a sharp drop in agitator torque to point B followed by a rise in torque to point C and another dip to point D. Then the torque rose to a peak at E, which in the previous series of tests seemed to mark the point when all of the coal particles were incorporated in spherical agglomerates. However, a microscopic examination of the suspension following run 27, which is represented in Figure 28, showed the material to be incompletely agglomerated at E. Therefore, it became apparent that point E could not be relied upon completely as a marker for agglomeration. In subsequent series of agglomeration tests, monitoring of agitator torque was supplemented by frequent sampling of the coal suspension and examination of the samples with a microscope to determine the point when all coal particles were incorporated in agglomerates.

Runs 27 and 24 represented by Figures 28 and 29, respectively, were part of a set of four runs made under the same conditions except for treatment time following the introduction of air. Run 27 with a treatment time of 10 min. was the shortest, whereas Run 24 with a treatment time of 65 min. was the longest. Run 26 with a treatment time of 25 min. and run 25 with a treatment time of 45 min. were the other runs in the set. For all of these runs the variation in agitator torque was similar between point A and point E. However, the structure and appearance of the product at the end of each run depended on the length of treatment. After a 10 min. treatment the product consisted of a mixture of flocs and smaller agglomerates, whereas after a 25 min. treatment the product consisted entirely of compact spherical agglomerates having a diameter of 0.15-0.20 mm. When the treatment time was increased to 45 min., the agglomerate size increased to 0.20 - 0.30 mm. A further increase in treatment time to 65 min. produced 2.0 - 3.5 mm diameter agglomerates composed of clusters of smaller agglomerates. The formation and breakup of clusters may have been responsible for the series of large swings in agitator

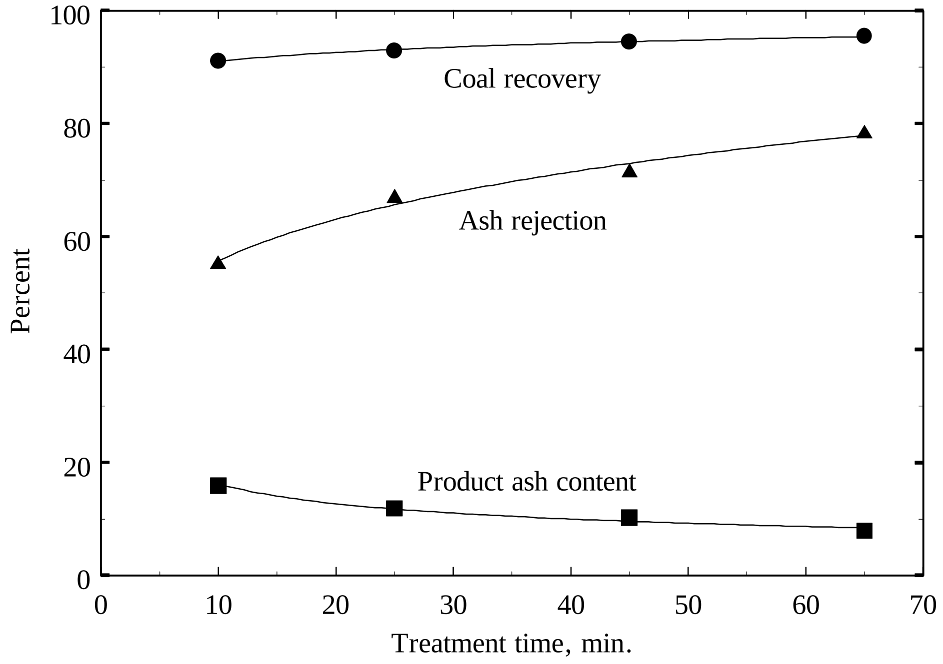


Figure 30. Effect of treatment time on coal recovery, ash rejection, and product ash content for runs 24 to 27.

torque observed near the end of run 24 (Figure 29). Similar swings in agitator torque were also observed in other runs which produced large agglomerates made up of smaller agglomerates.

An analysis of the product and tailings from runs 24 to 27 produced the results shown in Figure 30. It can be seen that as the treatment time was extended the coal recovery on a dry, ash-free basis increased gradually and the product ash content decreased. In addition, a larger percentage of the ash-forming mineral matter in the feed was rejected in the tailings. Therefore, extending the treatment time improved the separation efficiency of the process. The improvement was probably the result of producing larger agglomerates which were easier to separate by screening. For the shortest treatment time, agglomeration was incomplete which very likely accounted for the correspondingly poor separation. As the treatment time increased the agglomerate size also increased.

An examination of the results presented in Table 6 reveals a wide variation in agglomerate size, product ash content, ash rejection, and coal recovery depending on experimental conditions. The results appear to have been influenced strongly by both oil (i-octane) concentration and treatment time. The agglomerates tended to be larger, cleaner, and recovered in greater amount when an oil concentration of 30 v/w% was used, rather than 20 v/w%. Also the results tended to improve with treatment time for otherwise similar conditions.

The effects of treatment time and oil concentration on agglomerate growth is indicated by the results presented in Figures 31 and 32 for runs made with impeller speeds of 1750 and 1450 rpm, respectively. To achieve these results, the 11.43 cm tank was employed together with a solids concentration of 30 w/w% and an air concentration of 9 v/w%. It is apparent that agglomerate growth was much more rapid with 30 v/w% oil than with 20 v/w% oil regardless of impeller speed. Furthermore, for an oil concentration of 30 v/w% the growth was more rapid with an impeller speed of 1750 rpm than with a speed of 1450 rpm until a time was reached when large clusters were produced at the lower speed. Such clusters did not appear at the higher speed. The difference in agitator speed appeared to have little effect on agglomerate growth rate when an oil concentration of 20 v/w% was employed.

A direct indication of agglomerate size on product ash content, coal recovery, and ash rejection was obtained for runs 62 to 69. Such an indication was possible because the mean agglomerate diameter was determined for these runs by utilizing image analysis. The results presented in Figure 33 indicate that coal recovery increased markedly with increasing agglomerate diameter until a mean diameter somewhat less than 190 μm was produced. An increase in mean diameter beyond this value had only a small effect. Since the agglomerates were recovered with a screen having 250 μm apertures, the smaller agglomerates in any given product would not have been recovered.

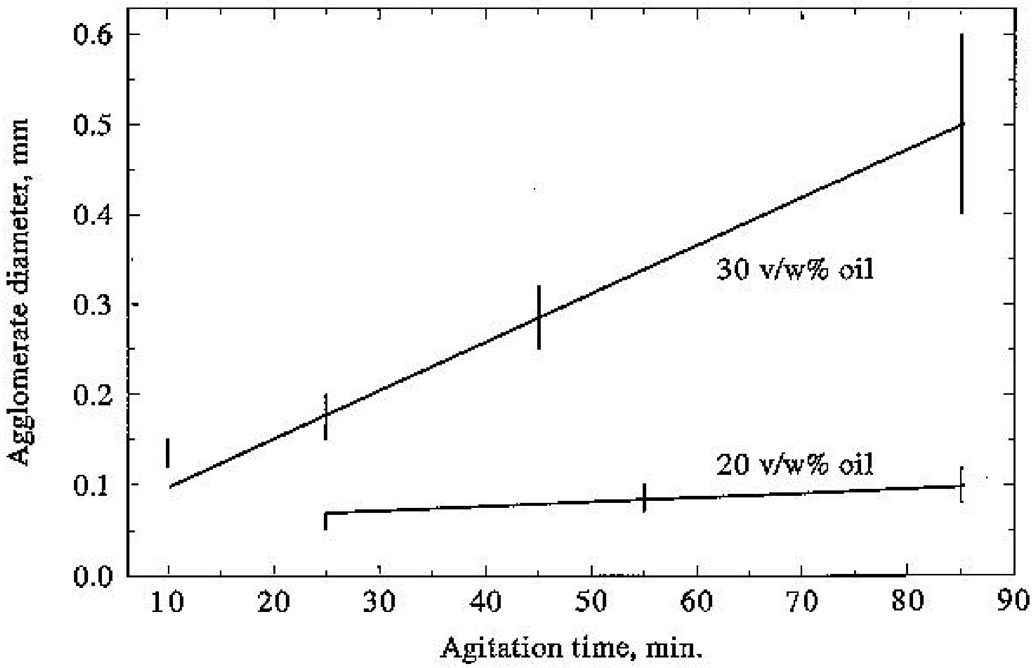


Figure 31. Variation in agglomerate size with treatment time for runs made with an agitator speed of 1750 rpm.

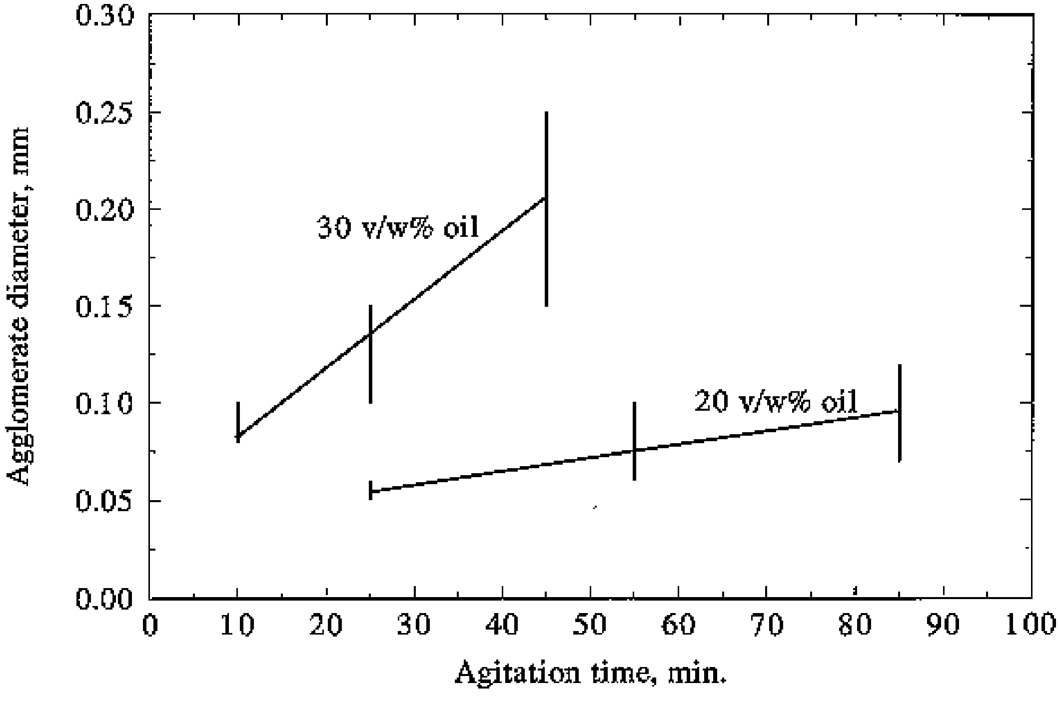


Figure 32. Variation in agglomerate size with treatment time for runs made with an agitator speed of 1450 rpm.

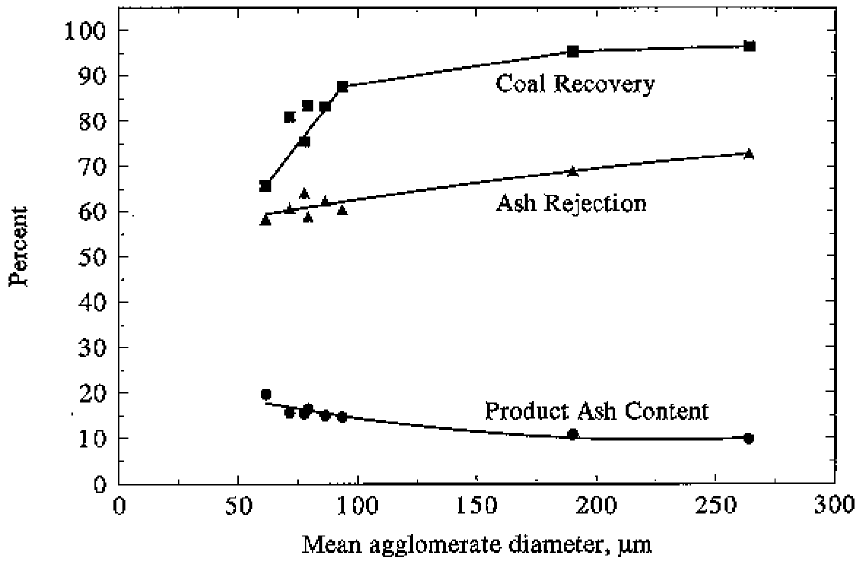


Figure 33. Effect of mean agglomerate diameter determined by image analysis on coal recovery, ash rejection, and product ash content for runs 62 to 69.

The results presented in Figure 33 also indicate that more ash was rejected resulting in a cleaner product as the mean agglomerate size rose. This trend was probably due to a decrease in the number of smaller agglomerates which because of their relatively large surface area were more likely to trap ash-forming mineral particles.

Although the results presented in Table 6 do not provide a clear indication of the effect of air concentration on agglomerate growth rate, they do show that coal recovery and ash rejection improved with increasing air concentration (see Figures 34 and 35). The better separation of agglomerates from mineral matter resulting from the use of more air suggests that the size distribution of the agglomerates was affected by air concentration even though the size range reported in Table 6 did not appear to vary with air concentration for runs made under comparable conditions. For runs 25, 43 and 47, which were the basis for Figure 34, the agglomerates were mainly in the 0.2 to 0.3 mm size range, while for runs 64, 72 and 80, which were the basis for

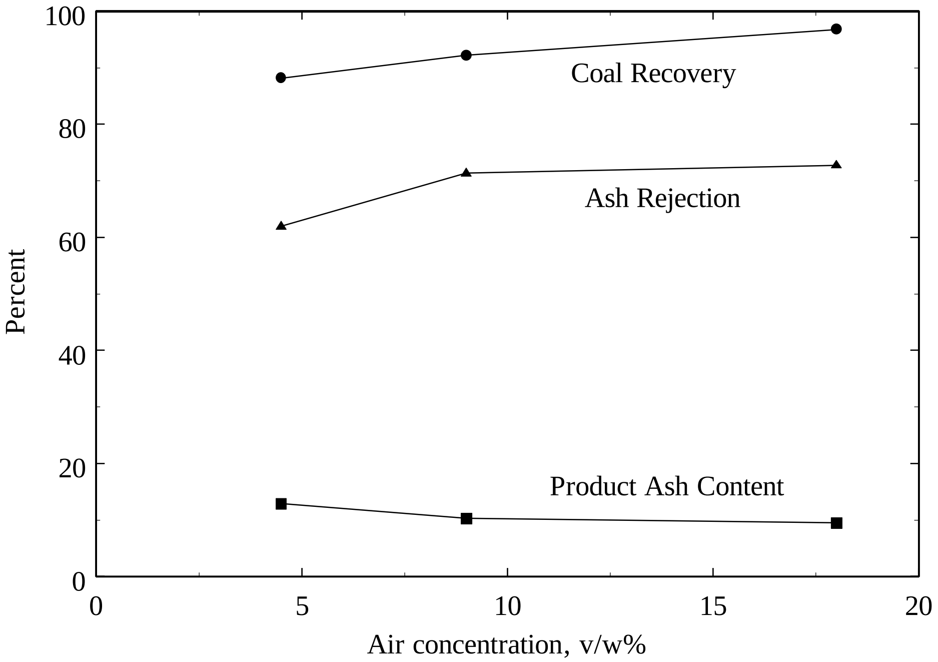


Figure 34. Effect of air concentration for runs made with 30 w/w% solids, 30 v/w% oil, and 45 min. treatment time using an agitator speed of 2000 rpm in 7.62 cm diameter tank.

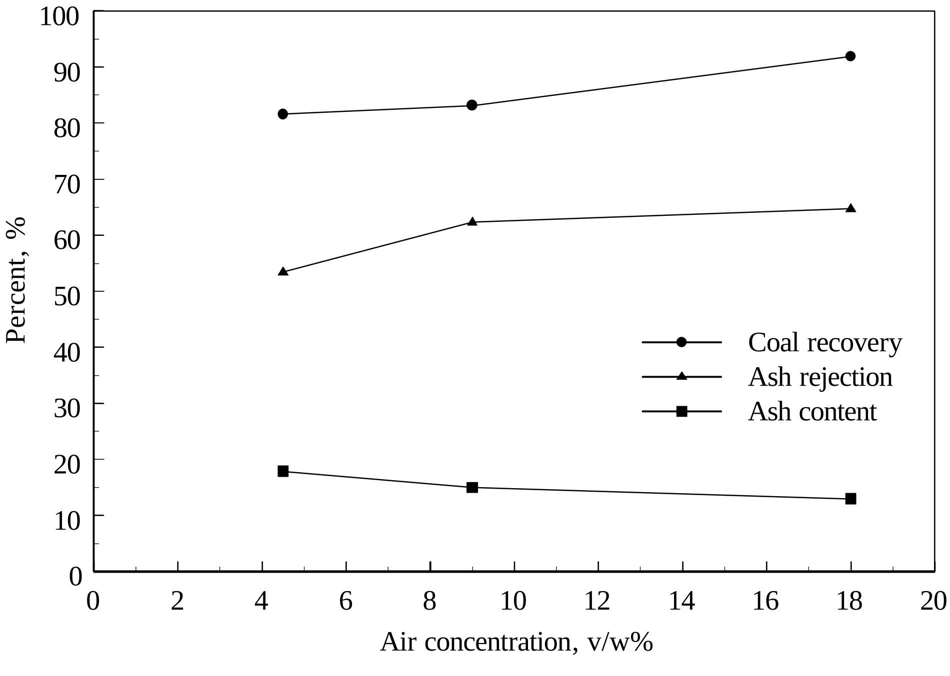


Figure 35. Effect of air concentration for runs made with 30 w/w% solids, 20 v/w% oil, and 85 min. treatment time using an agitator speed of 1600 rpm in 11.4 cm diameter tank.

Figure 35, the agglomerates were mainly in the 0.05 to 0.12 mm size range. The latter appeared to be considerably smaller than the screen aperture size (i.e., 0.25 mm) which illustrates the difficulty of separating small agglomerates by screening. Of course, the method used for measuring the range of agglomerate size provided only an approximate indication of the size distribution. A more accurate determination of the agglomerate size distribution by image analysis would have been much more likely to reveal differences in agglomerate size attributable to air concentration.

Principal Agglomeration Tests with Pittsburgh Coal

Coal from the Pittsburgh No. 8 seam was used for an extensive series of agglomeraiton tests which were designed to investigate the effects of different system parameters on the minimum time required to produce spherical agglomerates t_e, the size of the agglomerates d_p, coal recovery, and product ash content. For each agglomeration test, small samples of the coal suspension undergoing agglomeration were collected and examined frequently to augment the indication of agglomeration provided by changes in agitator torque. At the end of each run, a final sample of agglomerates was subjected to image analysis to determine the size distribution of the product.

The coal for this series of agglomeration tests was prepared by a somewhat different procedure than that used previously. Whereas previously a double roll crusher had been used to grind 5 mm size coal particles, a high-speed impact mill was used to grind the material for the present series of tests. In both cases, the material was ground further in a stirred ball mill, and the resulting suspension was partially dewatered to form a thick paste which was stored in a refrigerator at 5°C until used. Measurement of the particle size distributions by automated image analysis showed that the wet milled material used for the present tests had a projected area mean particle diameter of 3.86 μm with individual particles ranging from 1 to 16.6 μm in diameter. Therefore, the particles were slightly smaller than those used for the previous series of agglomeration tests.

For a majority of the tests, pure i-octane from the Burdick and Jackson Laboratories was used as an agglomerant. However, pesticide grade i-octane from Fisher Scientific was used in later tests in the series. This material had a purity of 99.5% according to the supplier.

Except for the determination of t_e and agglomerate size, the agglomeration tests were conducted largely as in the previous series. There were some minor changes which were not thought to affect either t_e or agglomerate size. These changes included the suspension of system degassing before the start of each run and the use of an improved agglomerate washing procedure. After run 95 the coal suspensions were no longer degassed, and starting with run 100 the agglomerates were washed by dumping the contents of the mixing tank into a deep vessel fitted with a 250 μm screen bottom which was immersed in a pail of water. The contents of the vessel were stirred gently, and eventually the screen was raised to recover the agglomerates. Starting with run 108 the procedure was modified further by using a hand-held wand equipped with water sprays to wash the agglomerates. The wand was used to stir and wash the agglomerates suspended in the vessel with the screen bottom. After 1 min. of washing, the screen was raised, and the agglomerates were allowed to drain for 1 min. The under side of the screen was then rinsed with a fine water spray, and the agglomerates were allowed to drain for 2 min. The agglomerates were subsequently transferred to a pan and dried. The tailings left in the pail were recovered by filtration and then dried. The weight and ash content of the products were determined.

While conducting this series of tests, it was observed that some particle flocculation took place during the initial conditioning period if an agitator speed of 2000 rpm or higher was employed. Therefore, starting with run 244 the conditioning agitator speed was limited to 1500 rpm. For runs made with an agitator speed less than 1500 rpm, the coal suspension was conditioned at the same speed as that used during the run. Starting with run 268, the conditioning speed was limited to 1000 rpm for runs made with the 24.0 cm diameter tank.

Another modification in procedure which was introduced starting with run 244 was the way in which air was introduced to start agglomeration. Whereas air had been introduced by withdrawing a measured amount of slurry from the mixing tank with a syringe, the modified procedure involved withdrawing slurry through a side outlet in the tank and collecting the slurry in a graduated cylinder. While the slurry was being withdrawn, the agitator speed was reduced to 400 rpm. The procedure required approximately 20 s to carry out.

The experimental conditions employed and the results obtained in this series of agglomeration tests or runs are presented in Table 7. The following independent parameters were varied among the tests: tank size, impeller diameter and speed, solids concentration, oil (i-octane) concentration, air concentration, and total run time. As a consequence of using different experimental conditions, there was a large variation in some dependent variables and less variation in others. It is apparent that there was a large variation in t_e and the mean agglomerate diameter d_p, a modest variation in the agglomerated product ash content, and little variation in the recovery of coal on a dry, ash-free basis. In most cases coal recovery was quite high, and it seemed to depend mainly on oil concentration. With a solids concentration of 20 to 30 w/w% and an oil concentration of 30 v/w%, coal recovery was generally above 95% on a dry, ash-free basis. Coal recovery declined somewhat with a lower oil concentration and/or higher solids concentration. Ash rejection was generally in the range of 60 to 80% which resulted in a product ash content of 7 to 10% in many cases. After the improved washing procedure was

Table 7. Experimental conditions and results for batch agglomeration tests with Pittsburgh No. 8 coal and i-octane: Principal Series.

Run No.	T, cm.	D, cm.	N, rpm	S, m/s	Solids, w/w %	Oil, v/w%	Air, v/w%	t_e min.	t_t min.	P/V,[a] W/L	Agglom. Size, mm		Ash,[b] w/w%	Ash[c] Rej., %	Recov.,[d] %
											range	mean			
90	15.24	5.08	1800	4.79	30	30	9	19	25	12.33	0.10-0.20	0.149	17.16	48.5	88.3
91	15.24	7.55	1063	4.24	30	30	9	6	25	25.33	0.10-0.20	0.195	12.28	63.1	96.8
92	15.24	6.35	1440	4.79	30	30	9	6	25	23.97	0.10-0.25	0.212	13.34	62.9	97.0
93	15.24	5.08	2089	5.55	30	30	9	6	25	18.72	0.10-0.20	0.204	16.93	48.5	94.9
94	15.24	6.35	1440	4.79	30	30	9	6	25	23.60	0.10-0.25	0.212	15.96	50.6	97.8
95	15.24	7.55	1200	4.79	30	30	9	3	15	37.03	0.10-0.20	0.153	13.60	58.5	97.2
100	11.43	3.65	1700	3.25	20	30	9	37	55	6.10	0.05-0.17	0.126	11.69	64.7	95.6
101	15.24	5.08	1300	3.46	20	30	9	51	55	4.60	0.04-0.18	0.123	14.51	54.0	96.6
102	11.43	3.65	2020	3.86	20	30	9	13	55	9.86	0.10-0.20	0.166	8.99	72.4	97.1
103	15.24	5.08	1900	5.05	20	30	9	11	55	13.49	0.12-0.22	0.175	9.33	71.8	96.9
104	15.24	5.08	1550	4.12	20	30	9	18	55	7.14	0.10-0.20	0.156	10.11	67.8	97.1
105	11.43	3.65	2400	4.59	20	30	9	9	55	13.90	0.11-0.22	0.176	8.50	74.2	96.8
106	24.00	7.55	1000	3.99	20	30	9	31	40	5.39	0.10-0.20	0.146	14.26	55.9	96.8
107	24.00	7.55	1150	4.59	20	30	9	15	40	8.05	0.08-0.20	0.147	12.04	63.5	97.0
108	15.24	5.08	1550	4.12	20	30	9	18.5	55	7.64	0.11-0.27	0.196	8.15	74.3	96.8
109	11.43	3.65	1850	3.54	20	30	9	17	55	7.54	0.12-0.22	0.177	7.89	76.5	95.9
110	7.62	3.65	1550	2.96	30	30	9	37	40	14.48	--	--	12.60	61.8	93.9
111	15.24	7.55	1000	3.95	30	30	9	7	40	20.40	0.14-0.30	0.224	9.96	69.7	97.5
112	15.24	7.55	1150	4.55	30	30	9	4	40	33.26	0.20-0.30	0.301	8.39	74.5	97.3
113	7.62	3.65	2000	3.82	30	30	9	15	40	26.85	0.07-0.20	0.170	7.06	79.0	95.1
114	15.24	7.55	900	3.56	30	30	9	10	40	15.51	0.15-0.31	0.228	9.89	69.3	97.3
115	7.62	3.65	1700	3.25	30	30	9	24	40	18.22	0.10-0.30	0.185	6.97	79.6	94.4
116	7.62	3.65	1550	2.96	40	30	9	39	55	15.17	0.05-0.50	0.254	10.18	69.5	91.9
117	7.62	3.65	1700	3.25	35	30	9	22	55	18.65	0.10-5.00	0.509	8.48	73.7	95.5
118	7.62	3.65	2000	3.82	40	30	9	6	55	27.28	0.20-0.80	0.315	10.67	67.6	91.7
119	7.62	3.65	2000	3.82	35	30	9	13	55	28.29	0.10-1.10	0.326	7.91	--	--
120	7.62	3.65	1550	2.96	35	30	9	31	55	13.24	0.10-0.40	0.210	9.30	74.6	92.6
121	7.62	3.65	1700	3.25	40	30	9	15	55	19.41	0.10-0.60	0.292	8.74	75.0	82.9

Run	T,	D,	N,	S,	Solids,	Oil,	Air,	t_e	t_t	P/V,[a]	Agglom. Size, mm		Ash,[b]	Ash[c] Rej.,	Recov.,[d]
No.	cm.	cm.	rpm	m/s	w/w %	v/w%	v/w%	min.	min.	W/L	range	mean	w/w%	%	%
126	7.62	3.65	1700	3.25	30	30	9	16	55	20.63	3.00-4.00	3.500	8.04	75.8	96.2
127	7.62	3.65	1700	3.25	35	30	9	26	55	19.38	0.50-6.00	3.200	8.76	73.4	96.5
128	7.62	3.65	1700	3.25	40	30	9	11	55	15.41	Flocs	--	6.51	79.9	93.8
129	7.62	3.65	1550	2.96	20	30	9	16	55	15.12	0.14-0.56	0.360	4.02	90.9	93.4
131	7.62	3.65	2000	3.82	35	20	9	28	115	25.01	0.02-0.10	0.055	11.59	69.1	81.5
132	7.62	3.65	2000	3.82	30	20	9	22	55	27.22	0.02-0.10	0.051	10.12	73.4	80.2
133	7.62	3.65	1700	3.25	40	20	9	55	55	14.01	0.01-0.10	0.041	23.25	66.5	41.1
134	7.62	3.65	1700	3.25	20	30	9	11	55	19.39	0.10-0.60	0.202	7.21	78.6	93.9
135	7.62	3.65	1700	3.25	35	20	9	112	145	16.82	0.01-0.16	0.047	13.92	64.8	82.0
136	7.62	3.65	2000	3.82	40	20	9	26	55	23.35	0.05-0.10	0.068	16.43	55.7	83.6
137	7.62	3.65	2000	3.82	20	30	9	5	55	25.93	0.10-0.60	0.169	7.11	79.2	93.5
138	7.62	3.65	1550	2.96	30	20	9	55	65	12.35	0.03-0.10	0.044	9.84	77.3	77.5
139	7.62	3.65	1550	2.96	40	20	9	97	150	12.35	0.02-0.10	0.034	26.26	16.1	88.5
140	7.62	3.65	1700	3.25	30	20	9	53	65	17.52	0.02-0.10	0.041	16.51	61.1	72.7
141	7.62	3.65	1700	3.25	35	30	9	25	55	18.69	0.10-1.30	0.315	10.73	69.1	94.2
142	7.62	3.65	2000	3.82	30	30	9	7	40	30.16	0.50-4.00	2.500	10.73	70.9	93.7
143	7.62	3.65	1550	2.96	30	30	9	32	40	14.91	0.10-0.28	0.151	7.13	79.0	93.7
144	7.62	3.65	1550	2.96	35	30	9	30	55	13.20	0.20-0.30	0.214	8.51	75.7	94.5
145	7.62	3.65	1700	3.25	20	30	9	11	55	19.85	0.10-0.45	0.178	7.22	78.3	95.1
146	7.52	3.65	2000	3.82	35	30	9	11	55	26.66	0.20-0.80	0.568	8.24	75.5	92.9
147	7.62	3.65	1700	3.25	30	30	9	17	40	19.39	0.10-0.40	0.181	7.29	79.4	95.3
148	7.62	3.65	2400	4.59	40	20	9	15	55	36.20	0.10-0.30	0.156	8.66	75.6	89.3
149	7.62	3.65	2400	4.59	35	20	9	9	55	40.17	0.08-0.50	0.114	7.30	78.6	94.1
150	7.62	3.65	2400	4.59	30	20	9	6	55	39.73	0.06-0.50	0.183	7.47	79.9	93.3
151	7.62	3.65	2400	4.59	20	20	9	10	55	42.60	0.10-0.60	0.177	6.42	81.8	90.4
152	7.62	3.65	2000	3.83	20	30	9	11	55	26.30	0.10-1.10	0.256	7.04	78.7	95.1
153	7.62	3.65	2400	4.59	30	30	9	2	55	46.58	0.30-0.40	0.360	7.42	77.6	96.0
154	7.62	3.65	2000	3.82	20	20	9	19	55	25.19	0.04-0.10	0.051	6.46	94.0	30.2
155	7.62	3.65	1700	3.25	25	30	9	10	55	18.22	2.50-4.00	3.200	7.43	78.0	96.0
156	7.62	3.65	2400	4.59	40	30	9	6.2	55	51.66	1.00-2.00	1.500	10.74	67.5	92.6
157	7.62	3.65	1550	2.96	20	20	9	31	55	12.14	0.03-0.15	0.059	6.50	91.6	41.1
158	7.62	3.65	1550	2.96	20	30	9	15	55	12.78	0.10-0.60	0.172	7.15	80.1	94.8
159	7.62	3.65	2000	3.82	25	30	9	8	55	28.51	0.20-0.50	0.302	7.54	77.3	94.6

Run No.	T, cm.	D, cm.	N, rpm	S, m/s	Solids, w/w %	Oil, v/w%	Air, v/w%	t_e min.	t_t min.	P/V,[a] W/L	Agglom. Size, mm range	mean	Ash,[b] w/w%	Ash[c] Rej., %	Recov.,[d] %
160	7.62	3.65	1700	3.25	20	20	9.0	25	55	17.05	0.03-0.10	0.050	5.79	98.0	10.8
161	7.62	3.65	1500	2.96	25	30	9.0	11	55	14.27	0.05-0.40	0.177	7.31	78.9	95.1
162	7.62	3.65	2400	4.59	35	30	9.0	6	55	33.32	0.15-1.00	0.321	7.73	79.9	88.2
163	7.62	3.65	1700	3.25	30	20	4.5	37	55	15.88	0.03-0.10	0.056	9.12	79.0	73.8
164	7.62	3.65	1550	2.96	20	20	18.0	17	55	12.99	0.05-0.15	0.073	6.77	84.5	75.3
165	7.62	3.65	2000	3.82	30	30	4.5	7.5	55	28.14	3.00-4.00	3.500	7.93	68.7	96.9
166	7.62	2.65	2000	3.82	30	20	18.0	15	55	24.27	0.05-0.30	0.132	6.82	81.4	87.8
167	7.62	3.65	1700	3.25	30	30	18.0	11	55	16.35	0.10-4.00	2.000	8.29	74.8	96.1
168	7.62	3.65	1550	2.96	30	25	9.0	31	55	12.35	0.10-0.22	0.107	7.77	78.5	94.7
169	7.62	3.65	1700	3.25	30	20	18.0	19	55	15.88	0.03-0.15	0.062	7.39	90.0	87.7
170	7.62	3.65	1550	2.96	30	20	18.0	35	55	11.92	0.03-0.15	0.045	13.56	63.3	88.1
171	7.62	3.65	2000	3.82	20	20	18.0	6	55	25.74	0.05-0.15	0.124	6.42	83.1	83.8
172	7.62	3.65	1550	2.96	30	30	4.5	23	55	11.92	0.10-0.60	0.196	7.52	77.6	95.3
173	7.62	3.65	2000	3.82	30	30	18.0	4.5	55	26.30	0.10-0.60	0.232	7.46	82.8	91.5
174	7.62	3.65	1700	3.25	30	25	9.0	15	55	15.88	0.10-0.22	0.224	7.20	79.8	94.9
175	7.62	3.65	2000	3.82	30	20	4.5	17	55	26.48	0.03-0.10	0.055	6.36	85.5	77.5
176	7.62	3.65	1700	3.25	20	20	18.0	13	55	16.82	0.03-0.13	0.051	5.98	90.1	53.3
177	7.62	3.65	1550	2.96	30	30	18.0	14	55	12.78	0.10-0.90	0.176	6.07	85.7	93.1
178	7.62	3.65	2000	3.82	30	25	9.0	8	55	25.74	0.10-0.60	0.208	7.11	79.1	95.3
179	7.62	3.65	1700	3.25	30	30	4.5	19	55	16.82	0.10-1.00	0.216	7.96	77.2	95.3
188	7.62	3.65	1550	2.96	30	30	9.0	23	40	15.33	0.20-0.35	0.217	7.42	78.4	92.2
189	7.62	3.65	1700	3.25	30	30	9.0	13	40	18.69	1.00-4.00	2.500	8.30	75.1	96.6
190	7.62	3.65	2000	3.82	40	30	9.0	17	55	19.78	0.80-1.50	1.000	7.57	80.7	85.3
191	7.62	3.65	1700	3.25	35	20	9.0	79	95	14.95	0.01-0.12	0.048	17.27	48.5	85.6
192	7.62	3.65	1700	3.25	30	20	9.0	21	55	15.88	0.03-0.15	0.054	8.71	74.6	91.7
193	7.62	3.65	1550	2.96	35	30	9.0	28	55	13.20	0.50-5.00	2.000	8.64	73.7	95.8
196	7.62	3.65	2000	3.82	30	30	4.5	5.5	55	30.53	1.00-2.20	1.500	7.71	77.3	94.4
197	7.62	3.65	2000	3.82	30	20	4.5	13.5	55	26.11	0.04.0.11	0.059	9.02	73.4	91.7
198	7.62	3.65	2000	3.82	35	30	9.0	13	55	25.74	3.50-4.50	4.000	8.13	76.7	96.2
199	7.62	3.65	2400	4.59	30	30	9.0	2.3	55	50.34	0.25-0.50	0.412	7.59	76.8	95.8
200	7.62	3.65	1550	2.96	35	20	9.0	163	55	13.20	0.02-0.12	0.060	17.67	43.1	92.4
201	11.43	3.65	2020	3.86	20	30	9.0	17	55	8.93	0.10-0.25	0.175	7.76	78.6	96.5
202	11.45	3.65	1850	3.54	20	30	9.0	17	55	7.19	0.10-0.20	0.146	7.47	78.0	96.2

Run No.	T, cm.	D, cm.	N, rpm	S, m/s	Solids, w/w %	Oil, v/w%	Air, v/w%	t_e min.	t_t min.	P/V,[a] W/L	Agglom. Size, mm		Ash,[b] w/w%	Ash[c] Rej., %	Recov.,[d] %
											range	mean			
203	15.24	5.08	1300	3.46	20	30	9.0	33	55	4.59	0.10-0.25	0.160	8.17	76.6	96.6
204	15.24	5.08	1550	4.12	35	30	9.0	48.5	55	7.29	0.10-0.25	0.122	17.18	46.4	95.5
205	2400	7.55	1000	3.95	20	30	9.0	25	40	5.55	0.10-0.25	0.143	8.33	76.3	96.1
208	7.62	3.65	2000	3.82	30	15	9.0	13.4	55	25.74	0.02-0.10	0.039	9.25	83.8	54.3
209	7.62	3.54	2000	3.82	30	25	9.0	9	55	29.06	0.12-0.35	0.205	7.72	76.7	96.6
210	7.62	3.65	2000	3.82	30	35	9.0	6	55	27.03	1.00-2.00	1.500	7.62	77.0	96.5
236	24.00	7.55	1000	3.95	30	30	9.0	19	55	5.65	0.10-0.40	0.109	11.90	65.1	97.2
237	11.43	5.08	1700	4.52	30	30	9.0	4	55	23.08	0.20-0.50	0.337	7.88	77.6	97.0
238	11.43	5.08	2000	5.32	30	30	4.5	3.2	55	41.54	0.25-0.46	0.367	6.46	80.0	97.5
239	24.00	7.55	1150	4.55	30	30	9.0	11	55	8.94	0.15-0.40	0.282	8.52	76.7	97.3
240	11.43	5.08	1700	4.52	30	30	18.0	5	55	23.62	0.20-0.50	0.359	8.08	77.4	96.9
241	11.43	5.08	1550	4.12	25	30	4.5	8.5	55	17.30	0.15-0.30	0.179	8.88	73.2	96.0
242	11.43	5.08	1700	4.52	30	25	9.0	5.5	55	24.08	0.20-0.30	0.212	8.07	77.2	96.6
243	11.43	5.08	1700	4.52	30	25	4.5	7.5	55	24.72	0.15-0.30	0.202	8.45	76.0	96.0
244	11.43	5.08	2000	5.32	30	30	9.0	2.5	55	40.03	0.30-0.60	0.476	8.50	75.7	97.0
245	11.43	5.08	1700	4.52	30	25	18.0	11	55	22.44	0.20-0.40	0.153	7.76	78.4	96.8
246	11.43	5.08	2000	5.32	30	25	4.5	5.5	55	39.71	0.10-0.25	0.178	8.79	75.4	96.2
247	11.43	5.08	2000	5.32	30	30	18.0	3	55	37.46	0.20-0.50	0.323	7.92	77.6	97.2
248	11.43	5.08	2000	5.32	30	25	9.0	3.5	55	39.39	0.20-0.30	0.216	8.52	76.2	96.9
249	11.43	5.08	2000	5.32	30	30	4.5	4	55	38.85	0.25-0.50	0.470	8.80	74.4	97.4
250	11.43	5.08	1700	4.52	30	30	4.5	6	55	23.99	0.20-0.50	0.314	8.68	75.0	97.3
254	11.43	5.08	2000	5.32	30	30	18.0	3	55	35.95	0.20-0.60	0.332	7.66	78.1	97.2
270	24.00	7.55	1200	4.74	30	30	9.0	13	55	7.88	0.15-0.40	0.284	7.62	78.4	97.2
271	24.00	7.55	1400	5.53	30	30	9.0	7	55	13.70	0.20-0.70	0.304	8.38	74.3	97.4
272	11.43	3.65	2200	4.20	30	30	9.0	10	55	11.70	0.10-0.40	0.241	7.43	78.9	96.2
273	11.43	3.65	2400	4.59	30	30	9.0	8	55	14.20	0.15-0.32	0.252	7.75	77.8	96.4
274	15.24	5.08	2000	5.32	30	30	9.0	5.5	55	15.80	0.20-0.35	0.268	8.25	76.4	97.0
275	24.00	7.55	1000	3.95	30	30	9.0	23	55	4.34	0.10-0.30	0.185	10.78	70.8	95.8
276	11.43	3.65	2000	3.82	30	30	9.0	15.5	55	8.15	0.10-0.30	0.235	7.62	77.5	96.1
277	15.24	5.08	1700	4.52	30	30	9.0	11	55	9.54	0.10-0.40	0.246	7.53	75.6	97.1
278	15.24	5.08	2200	5.85	30	30	9.0	5	55	22.10	0.10-0.40	0.280	7.38	77.6	97.1

Run No.	T, cm.	D, cm.	N, rpm	S, m/s	Solids, w/w %	Oil, v/w%	Air, v/w%	t_e min.	t_t min.	P/V,[a] W/L	Agglom. Size, mm range	mean	Ash,[b] w/w%	Ash[c] Rej., %	Recov.,[d] %
279	15.24	5.08	1550	4.12	30	30	9.0	18	55	6.95	0.10-0.32	0.232	8.03	76.8	96.7
280	15.24	5.08	2400	6.38	30	30	9.0	3.5	55	28.10	0.20-0.40	0.334	8.39	75.1	97.3
281	15.24	5.08	1700	4.52	30	30	9.0	9	55	9.58	0.15-0.37	0.262	8.79	72.1	96.5

[a] Power input at t_e
[b] Ash content of agglomerates
[c] Ash rejected to tailings
[d] Coal recovery on a dry, ash-free basis

instituted in run 100, a higher product ash content was generally associated with the formation of very small agglomerates.

The experimental results were analyzed by applying a combination of multiple linear regression analysis and analysis of variance to determine the dependency of each of the principal dependent parameters on the independent variables. An equation of the following form was fitted to the experimental data for each dependent parameter Y_i:

$$Y_i = aX_1^b X_2^c X_3^d T^e D^f N^g \tag{34}$$

where X_1 = fractional oil concentration, v/w%/100
X_2 = fractional air concentration, v/w%/100
X_3 = fractional solids concentration, w/w%/100
T = tank diameter, m
D = impeller diameter, m
N = impeller speed, rpm

Fitting was accomplished by converting the preceding equation to a linear form by taking the logarithm of both sides and then applying multiple linear regression analysis to determine the a…g coefficients. Values of the coefficients determined by this procedure are listed in Table 8 together with the corresponding multiple correlation coefficient R. The number of observations utilized for each fitting is also indicated. Fewer observations were utilized for fitting d_p than for fitting the other dependent parameters because only the results of runs which produced agglomerates smaller than 1.0 mm were included. The results of runs which produced large clusters of smaller agglomerates were omitted because such results seemed less reproducible and more difficult to correlate.

The statistical significance of the contribution made by each of the independent variables appearing in equation 34 was tested by conducting an analysis of variance. The resulting p-values are listed in Table 9. A p-value of 0.05 or less indicates a high level of significance.

Table 8. Values of the coefficients appearing in equation 34 determined by multiple linear regression analysis for Pittsburgh No. 8 coal and values of R for each correlation.

Independent variable	Coefficient	Dependent parameter (Y_i)			
		t_e, min.	$d_{p, mm}$	Ash, %	Recovery, %
[Intercept)	a	35.8 x 10^6	0.016	160.6	166.5
Oil conc. (X_1)	b	-1.963	3.291	-0.500	0.682
Air conc. (X_2)	c	-0.261	0.080	-0.047	0.024
Solids conc. (X_3)	d	1.018	0.194	0.658	0.264
Tank dia. (T)	e	2.983	-0.964	0.638	0.115
Imp. Dia. (D)	f	-6.798	1.995	-0.650	0.006
Speed (N)	g	-4.199	1.502	-0.458	0.125
Correl. Coef. (R)		0.902	0.907	0.573	0.551
No. observations		127	118	126	126

Table 9. Statistical p-values determiend by an analysis of variable of correlations for Pittsburgh No. 8 coal.

Independent variable	Dependent parameter (Y_i)			
	t_e	d_p	Ash	Recovery
[Intercept]	0.0001	0.0002	0.0001	0.0001
Oil conc. (X_1)	0.0001	0.0001	0.0005	0.0001
Air conc. (X_2)	0.0153	0.3716	0.5120	0.7056
Solids conc. (X_3)	0.0001	0.1681	0.0001	0.0110
Tank dia. (T)	0.0001	0.0001	0.0003	0.4505
Imp. dia. (D)	0.0001	0.0001	0.0180	0.9804
Speed (N)	0.0001	0.0001	0.0022	0.3312

The results of the preceding analyses show that a good correlation was obtained between t_e and the six independent variables since the multiple correlation coefficient was high (R = 0.902). Furthermore, each of the independent variables made a statistically significant contribution to t_e since the corresponding p-values were all less than 0.05. The results also indicate a good correlation between agglomerate diameter (d_p) and the independent variables because the multiple correlation coefficient was again large (R = 9.07). However, in this case neither air concentration nor solids concentration made a statistically significant contribution to d_p because the corresponding p-values were greater than 0.05. The correlation between the product ash content and the independent variables was not as good as the preceding correlations since the multiple correlation coefficient was much lower (R = 0.573). The large p-value for air concentration shows that this variable did not make a statistically significant contribution to the ash content, but the ash content did appear to depend on all of the other variables. Finally, it can be seen that the correlation coefficient for coal recovery (0.551) was also low and that only oil concentration and solids concentration made a significant contribution to coal recovery.

An attempt was made to find better correlations for both t_e and d_p than the correlations based on equation 34. A number of different empirical models were tested including models based on dimensional analysis. In the case of t_e, the following correlation proved to be the best since it had the highest correlation coefficient (R = 0.968) of any models tested:

$$\frac{t_e}{V} = 107.8\ X_1^{-1.605} X_2^{-0.274} X_3^{0.859} P^{-1.354} \qquad (35)$$

where P = agitator power, kW
V = tank volume, liters

All of the independent variables in this model made a statistically significant contribution to t_e/V since the corresponding p-values were all less than 0.05. The correlation coefficient was appreciably higher than for a correlation based on dimensional analysis for which R was 0.831.

In the case of d_p, a correlation was not found which fit the data better than the correlation based on equation 34. However, the following simpler equation fit the data almost as well since it had a correlation coefficient of 0.904:

$$d_p = 1.349\, X_1^{3.138}\, (D/T)^{-0.541}\, (P/V)^{0.555} \tag{36}$$

Furthermore, each of the variable terms on the right-hand side of the equation made a statistically significant contribution to d_p.

The results represented by equation 35 show that for Pittsburgh No. 8 coal, t_e increases with increasing solids concentration and suspension volume or, in other words, with the amount of coal present when other conditions are held constant. By the same token t_e decreases with increasing concentrations of i-octane and air or increasing agitator power. Therefore, t_e can be reduced by increasing any of these three parameters up to a limit which has not been established.

Similarly equation 36 shows that d_p depends very strongly on i-octane concentration but not at all on the concentrations of solids or air. Therefore, a small increase in i-octane concentration will produce a large increase in d_p. It is also apparent that d_p will increase with an increase in P/V or a decrease in D/T, but the effects are not as strong as the effect of i-octane concentration.

While conducting the agglomeration tests, it was observed that an i-octane concentration of 35 v/w% was an upper practical limit because above this concentration much of the coal tended to stick to the walls of the mixing tank.

Agglomeration Tests With Upper Freeport Coal

Coal from the Upper Freeport Seam was used for a limited series of agglomeration tests which were designed to investigate the effects of different system variables on t_e, d_p, coal recovery, and product ash content. The coal for this series of tests had an ash content of 25.5 wt.%. It was prepared by passing it through a jaw crusher and a high-speed impact mill. Then it was ground in a stirred ball mill as previously described, and after partial dewatering, the resulting paste was stored in a refrigerator at 5°C until needed. The wet milled coal had a projected area mean particle diameter of 4.72 μm determined by automated image analysis. Therefore, the mean diameter was only slightly smaller than that for the Pittsburgh No. 8 coal used for the previous series of tests.

Pure i-octane from the Burdick and Jackson Laboratories was used as an agglomerant for the earlier tests in the series, while pesticide grade i-octane (99.5%) was used as an agglomerant for the later tests.

The same experimental procedure was employed for this series of tests as for the previous series which meant that the changes in procedure incorporated in the previous series were incorporated in this series as well. Work on the present series was started before the previous series was completed, and since a single series of run numbers was used for both series of tests, the order in which the tests were conducted is indicated by run number regardless of series.

The experimental conditions employed and the results achieved with Upper Freeport coal are indicated in Table 10. As in the case of Pittsburgh No. 8 coal, the following independent parameters were varied among tests: tank size, impeller diameter and speed, solids concentration, oil (i-octane) concentration, and air concentration. As a consequence of the variation in experimental conditions, there was a large variation in both t_e and d_p but only a small

Table 10. Experimental conditions and results of oil agglomeration runs made with Upper Freeport coal and i-octane

Run, No.	T, cm	D, cm	N, rpm	S, m/s	Solids, w/w%	Oil, v/w%	Air, v/w%	t_e min	t_t min	P/V, [a] kW/m^3	Aggl. Size, mm range	Aggl. Size, mm mean	Ash,[b] w/w%	Ash[c] Rej.,%	Recov.,[d] %
180	7.62	3.65	2000	3.82	20	20	9.0	45	55	25.01	0.02-0.08	0.035	- - -	- - -	- - -
181	7.62	3.65	2000	3.82	30	30	9.0	4.3	55	26.48	1.00-1.20	1.500	9.36	69.8	96.9
182	7.62	3.65	2000	3.82	20	30	9.0	5	55	26.11	1.00-2.00	1.500	8.63	72.9	94.9
183	7.62	3.65	1700	3.25	30	20	4.5	90	55	17.03	0.03-0.10	0.054	20.26	51.1	65.8
184	7.62	3.65	1700	3.25	30	30	9.0	7	55	16.82	1.00-2.00	1.500	8.88	72.2	96.5
185	7.62	3.65	1700	3.25	20	30	9.0	4	55	17.05	0.50-2.50	1.400	8.67	73.0	95.6
186	7.62	3.65	1550	2.96	30	30	9.0	9	55	12.78	1.00-3.50	2.000	9.10	71.7	96.0
187	7.62	3.65	1550	2.96	20	30	9.0	5	55	13.41	0.05-0.30	- - -	8.40	75.2	92.2
206	7.62	3.65	2000	3.82	30	20	9.0	51	65	25.38	0.03-0.08	0.049	- - -	- - -	- - -
207	7.62	3.65	2000	3.82	30	20	18.0	16	55	25.93	0.03-0.11	0.058	10.09	79.4	61.7
220	7.62	3.65	2200	4.20	30	20	4.5	32	55	32.17	0.01-0.10	0.048	- - -	- - -	- - -
221	7.62	3.65	2200	4.20	30	20	18.0	15	55	22.37	0.01-0.07	0.046	- - -	- - -	- - -
222	7.62	3.65	2400	4.59	30	20	18.0	9	55	29.12	0.01-0.06	0.042	- - -	- - -	- - -
223	7.62	3.65	2000	3.82	30	10	18.0	53	55	22.81	0.01-0.07	0.020	- - -	- - -	- - -
224	7.62	3.65	2400	4.59	30	10	40.0	31	55	30.89	0.01-0.10	0.020	- - -	- - -	- - -
225	7.62	3.65	2000	3.82	30	25	9.0	5	85	26.48	0.05-0.70	0.133	9.58	72.1	93.4
226	7.62	3.65	1700	3.25	30	25	9.0	6	55	16.35	0.05-0.50	0.103	9.42	72.2	94.6
227	7.62	3.65	1550	2.96	30	25	9.0	11	55	12.99	0.05-0.35	0.108	9.34	72.4	95.2
228	11.43	3.65	1700	3.25	30	30	9.0	15	55	5.38	0.15-0.31	0.227	11.77	65.9	94.7
229	15.24	5.08	1700	4.52	30	30	9.0	5	55	9.77	0.12-0.24	0.263	11.40	64.8	97.2
230	11.43	3.65	2400	4.59	30	30	9.0	3	55	12.68	0.15-0.25	0.184	12.15	61.5	94.9
231	11.43	3.65	2000	3.82	30	30	9.0	6	55	7.45	0.10-0.30	0.165	9.85	70.5	96.1
232	15.24	5.08	2000	5.32	30	30	9.0	3.5	55	15.40	0.15-0.25	0.251	10.02	69.5	97.2
233	15.24	5.08	1550	4.12	30	30	9.0	8.6	55	6.95	0.15-0.30	0.217	11.81	62.2	97.1
234	24.00	7.55	1000	3.95	30	25	9.0	15	55	5.68	0.06-0.13	0.098	18.17	39.3	97.1
235	24.00	7.55	1150	4.55	30	25	9.0	8	47	8.71	0.10-0.22	0.167	13.11	59.2	96.5
251	11.43	5.08	1550	4.12	30	30	4.5	9.5	55	17.05	0.20-0.30	0.213	9.90	70.0	96.1
252	11.43	3.65	1700	3.25	30	30	4.5	15	55	4.83	0.10-0.30	0.205	10.32	69.5	94.0
253	11.43	3.65	1550	2.96	30	30	4.5	111	115	4.57	0.10-0.30	0.111	9.30	72.4	95.2
255	11.43	3.65	2000	3.82	25	30	4.5	16	55	7.93	0.10-0.20	0.129	9.21	72.4	94.7
256	11.43	3.65	1700	3.25	25	30	22.0	10	55	4.92	0.10-0.32	0.150	8.62	75.1	94.9
257	11.43	3.65	1700	3.25	25	30	18.0	10	55	5.24	0.10-0.30	0.164	8.64	74.3	95.2

Table 10. Continued

Run, No.	T, cm	D, cm	N, rpm	S, m/s	Solids, w/w%	Oil, v/w%	Air, v/w%	t_e min	t_t min	P/V[a] kW/m^3	Aggl. Size, mm		Ash,[b] w/w%	Ash[c] Rej.,%	Recov.,[d] %
											range	mean			
258	11.43	3.65	1700	3.25	30	30	4.5	39	55	5.19	0.05-0.20	0.100	11.15	65.7	94.9
259	11.43	3.65	1550	2.96	30	30	18.0	22	55	3.99	0.10-0.30	0.181	16.37	47.5	94.5
260	11.43	3.65	1700	3.25	25	30	4.5	27	55	5.83	0.10-0.20	0.137	9.21	72.4	95.5
261	11.43	3.65	2000	3.82	25	30	18.0	5.5	55	8.04	0.10-0.21	0.172	8.61	74.4	95.4
262	11.43	3.65	2000	3.82	30	30	18.0	5	55	7.29	0.15-0.30	0.242	8.37	76.2	93.1
263	11.43	3.65	2000	3.82	30	30	4.5	9	55	7.72	0.10-0.30	0.190	8.96	74.10	94.7
264	11.43	3.65	1550	2.96	25	30	18.0	12	55	4.15	0.10-0.30	0.157	8.42	75.6	95.1
265	11.43	3.65	1550	2.96	25	30	4.5	40	55	4.57	0.06-0.15	0.115	7.95	77.6	93.9
266	15.24	5.08	1700	3.25	30	30	9.0	9.5	55	9.99	0.08-0.15	- - -	8.69	73.9	97.0
267	24.00	7.55	1500	5.93	30	30	9.0	5	15	15.93	0.10-0.20	- - -	10.21	68.2	96.6
268	24.00	7.55	1200	4.74	30	30	9.0	13	55	7.80	0.06-0.22	0.166	9.33	72.2	96.5
269	24.00	7.55	1000	3.95	30	30	9.0	27	55	4.62	0.10-0.22	0.157	10.42	69.0	96.3
282	15.24	5.08	2000	5.32	30	30	9.0	6	55	15.62	0.05-0.27	0.174	9.74	70.4	97.2
283	11.43	3.65	1700	3.25	30	30	9.0	20	55	5.19	0.11-0.25	0.168	9.38	71.1	95.1
284	15.24	5.08	2200	5.85	30	30	9.0	4	55	24.61	0.13-0.32	0.228	9.94	70.4	96.7

[a] Power input at t_e

[b] Ash content of agglomerates

[c] Ash rejected to tailings

[d] Coal recovery on a dry, ash-free basis

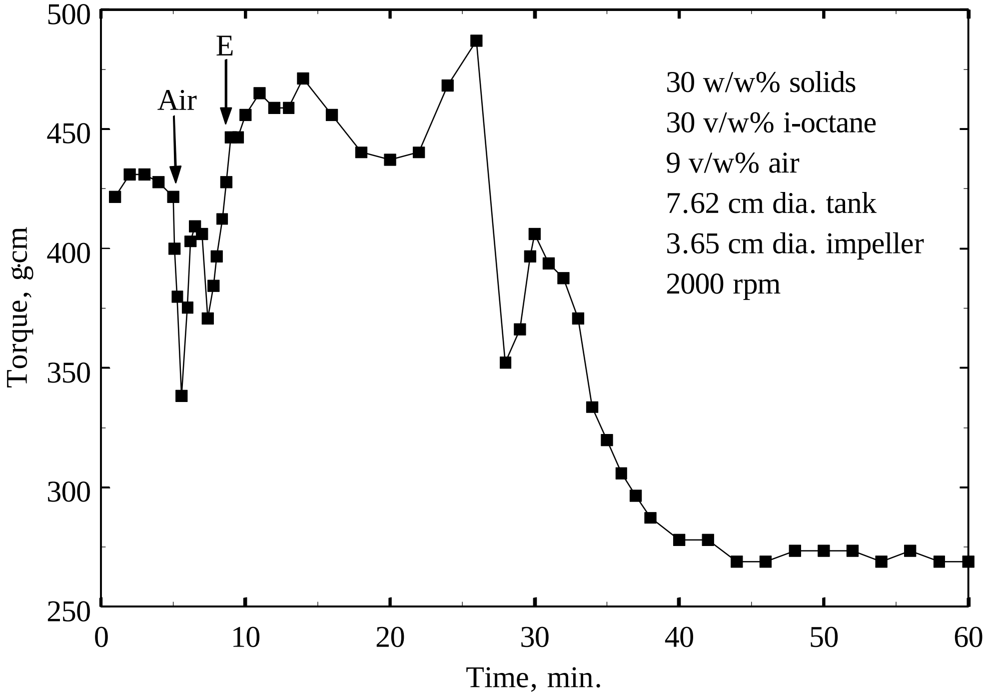


Figure 36. Changes in agitator torque for run 181 made with Upper Freeport coal.

variation in product recovery and ash content. In a large majority of runs, coal recovery was 95% or more and the ash content of the product was in the range of 8 to 12%. In almost all cases from 60 to 80% of the ash was rejected to the tailings.

As in the previous series, the progress of agglomeration was monitored by observing changes in agitator torque supplemented by microscopic examination of the suspension. The variation in torque with time is indicated by Figure 36 for run 181 which was conducted under conditions similar to those employed for run 24 with Pittsburgh No. 8 coal resulting in the torque readings presented in Figure 29. A comparison of these two figures reveals both similarities and differences. In both cases after air was introduced, the torque dropped, then rose to an intermediate peak, and dropped again. The torque then rose as spherical agglomerates were produced. By the time point E was reached most of the coal particles had been incorporated in agglomerates. In run 24 point E corresponded to the peak torque whereas in run 181 point E was identified by microscopic examination of samples of the suspension undergoing agglomeration. For run 24 the value of t_e was 7.5 min., whereas for run 181 the value of t_e was 4.3 min. which shows that the rate of agglomeration was faster for Upper Freeport coal than for Pittsburgh No. 8 coal.

The large swings in agitator torque which started occurring approximately 17 min. after air was introduced in run 181 seemed to be associated with the formation and breakage of large clusters of agglomerates because the final product consisted mostly of 1.5 mm diameter agglomerates made up of smaller agglomerates. A somewhat similar series of swings in agitator torque which started about 54 min. after air was introduced in run 24 was also associated with the formation and breakage of large clusters. Again it is apparent that the progress of important events was more rapid with Upper Freeport coal than with Pittsburgh No. 8 coal.

As in the case of Pittsburgh No. 8 coal, the results obtained with Upper Freeport coal were analyzed by fitting equation 34 to the data by employing multiple linear regression. Values of the coefficients determined by this method are listed in Table 11 together with the corresponding multiple correlation coefficient R. As before in developing a correlation for d_p, only those runs were included which produced agglomerates smaller than 1.0 mm. Also as before the statistical significance of the contribution made by each independent variable appearing in equation 34 was tested by calculating the corresponding p-value. The resulting p-values are listed in Table 12.

In the case of t_e, equation 34 did not fit the experimental results for Upper Freeport coal as well as for Pittsburgh No. 8 coal, since the multiple correlation coefficient for Upper Freeport coal (0.836) was lower than that for Pittsburgh coal (0.902). In addition, solids concentration did not contribute significantly to the fit for Upper Freeport coal, whereas it did contribute

Table 11. Values of the coefficients appearing in equation 34 determined by multiple linear regression analysis for Upper Freeport coal and values of R for each correlation.

Independent variable	Coefficient	Dependent parameter (Yi) t_e	d_p	Ash, %	Recovery, %
[Intercept]	a	1362	0.705	10.91	584
Oil conc. (X_1)	b	-3.404	2.293	-1.068	0.774
Air conc. (X_2)	c	-0.803	0.131	-0.047	0.001
Solids conc. (X_3)	d	0.080	1.242	0.326	0.076
Tank dia. (T)	e	1.916	0.449	0.279	-0.055
Imp. dia. (D)	f	-4.108	-0.281	-0.234	0.093
Speed (N)	g	-2.666	0.432	-0.169	-0.080
Correl. coef. (R)		0.836	0.930	0.662	0.838
No observations		47	37	40	40

Table 12. Statistical p-values determined by an analysis of variance of correlations for Upper Freeport coal.

Independent variable	Dependent parameter (Yi) t_e	d_p	Ash, %	Recovery, %
[Intercept]	0.0606	0.8768	0.1132	0.0001
Oil conc. (X_1)	0.0001	0.0001	0.0018	0.0001
Air conc. (X_2)	0.0001	0.1677	0.4425	0.9658
Solids conc. (X_3)	0.9072	0.1035	0.2116	0.3607
Tank dia. (T)	0.0007	0.2555	0.1374	0.3589
Imp. dia. (D)	0.0001	0.5946	0.3722	0.2711
Speed (N)	0.0001	0.1587	0.3628	0.1840

significantly to the fit for Pittsburgh coal. On the other hand, in the case of either d_p, ash content, or coal recovery, equation 34 seemed to fit the results for Upper Freeport coal better than it fit the results for Pittsburgh Coal. Interestingly, oil concentration seemed to be the only independent variable which contributed significantly to the fit for either d_p, ash content, or coal recovery for Upper Freeport coal according to the p-values listed in Table 12.

No equations were found which fit the data for either t_e or d_p better than equation 34. However, by eliminating variables which did not contribute significantly to the fit, it was possible to obtain simpler equations. By eliminating the solids concentration from equation 34 and refitting the data for Upper Freeport coal the following simpler equation was obtained for t_e:

$$t_e = 1155\ X_1^{-3.42}\ X_2^{-0.805}\ T^{1.93}\ D^{-4.11}\ N^{-2.66} \tag{37}$$

Since the correlation coefficient for this equation was 0.835, the fit was nearly as good as that noted for equation 34. Also every independent variable contributed significantly to t_e.

By eliminating T, D, and N from equation 34 and refitting the data for Upper Freeport coal, the following simpler equation was obtained for d_p:

$$d_p = 20.8\ X_1^{2.45}\ X_2^{0.158}\ X_3^{1.214} \tag{38}$$

Since the correlation coefficient for this equation is 0.922, it fits the data nearly as well as equation 34 fits. However, X_2 and X_3 make a marginal contribution to the fit because the p-values for these variables are 0.072 and 0.095, respectively. In other words, these p-values are slightly greater than the value of 0.05 which is considered statistically significant. By further elimination of X_2 and X_3 from the correlation for d_p, the much simpler expression is obtained,

$$d_p = 2.33\ X_1^{2.21} \tag{39}$$

This expression still provides a relatively good representation of the data since it has a correlation coefficient of 0.909.

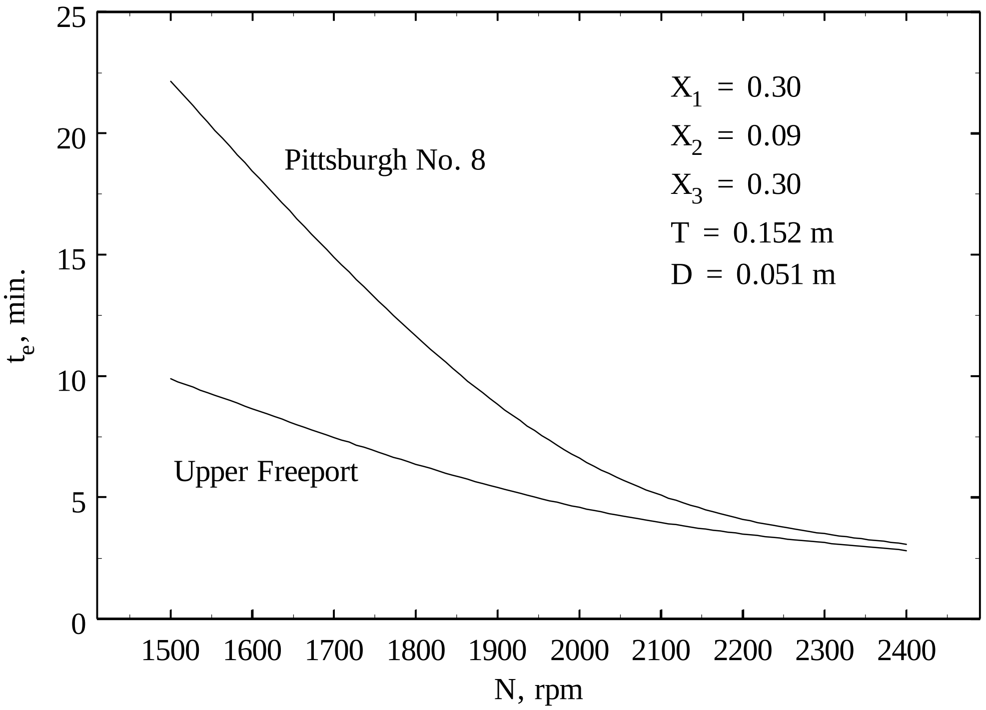


Figure 37. Effect of agitator speed predicted by equation 34 for two different coals.

Comparison of Agglomeration Characteristics

The correlations represented by equation 34 provide a means for comparing the agglomeration characteristics of Pittsburgh No. 8 coal and Upper Freeport coal. Consider the case represented by the following conditions: $X_1 = 0.30$, $X_2 = 0.09$, $X_3 = 0.30$, $T = 0.1524$ m, and $D = 0.0508$ m. Values of t_e given by equation 34 for each type of coal and these conditions are indicated in Figure 37 for various values of N. It can be seen that t_e is always greater for Pittsburgh No. 8 coal than for Upper Freeport coal which is probably due to the greater hydrophobicity of Upper Freeport coal. Also it can be seen that t_e decreases more rapidly with increasing agitator speed for Pittsburgh No. 8 coal than for Upper Freeport coal. Consequently at the highest speed t_e is almost the same for both coals.

It follows that the effects on t_e of changing other variables can be compared in a similar way for the two coals.

Size Scale-up of Mixing Systems

One of the objectives of the investigation was to determine a suitable basis for size scale-up of the mixing system used for agglomeration. The general approach for establishing such a basis is to find a key parameter or group of parameters which if kept constant, will produce the same results in systems of different size. Two parameters which have been widely used as a basis for scale-up of mixing systems are the power input per unit volume (P/V) and the impeller tip speed (13, 21). To see whether one or the other of these parameters would serve as a basis for scale-up of an oil agglomeration system, a group of runs was made with each type of coal using a series of tanks which differed in size. These runs were included in the preceding series of runs made with concentrated suspensions of Pittsburgh No. 8 coal and Upper Freeport coal. Conditions were chosen to preserve geometric similitude, i.e., the ratio of impeller diameter to tank diameter and the ratios of other critical dimensions were kept constant or nearly constant as different tanks in the series were employed. Also while focusing on size scale-up, the concentrations of solids, oil, and gas were kept constant along with the total time. The conditions common for all runs included 30 w/w% solids, 30 v/w% i-octane, 9 v/w% air, and 55 min. total treatment time. In addition, the ratio of impeller diameter to tank diameter was held in the range of 0.315 to 0.333 as tank size was varied. The tanks employed for this effort ranged in size from 11.43 cm I.D. to 24.0 cm I.D. The smallest available tank which had an inside diameter of 7.62 cm was not included because the results obtained with this tank did not fit the pattern provided by the other tanks. Several runs were made with each tank in order to vary both the agitator power input and impeller tip speed.

For Pittsburgh No. 8 coal, an almost perfect correlation was found between t_e/V and agitator power (see Figure 38). The equation shown below fits the data for all tank sizes extremely well because the multiple correlation coefficient is 0.997.

$$t_e = 151.1\ V/P^{1.095} \tag{40}$$

Since this equation represents the results obtained with mixing systems of various sizes, it suggests that a suitable basis for scale-up is to hold $V/P^{1.095}$ constant while, of course, maintaining constant geometric similitude and holding the concentrations of solids, oil, and air constant.

An alternative basis for scale-up bears consideration, and it involves correlating the results of the same set of runs with Pittsburgh No. 8 coal in terms of d_p instead of t_e. The best correlation found is between d_p and P/V and is represented by the following expression:

$$d_p = 152\ (P/V)^{0.222} \tag{41}$$

Since this equation has a correlation coefficient of 0.746, it does not fit the experimental data nearly as well as equation 40 fits the data. Part of the difficulty lies in the greater apparently random variation in d_p which can be seen in Figure 39. Therefore, holding $(P/V)^{0.22}$ constant while scaling up the size of the mixing system appears more doubtful than the preceding basis.

Since neither t_e nor d_p correlated well with impeller tip speed in the case of Pittsburgh No. 8 coal, the use of constant tip speed as a basis for system scale-up does not appear to be a useful option for a mixing system treating this coal. However, the results proved very different for systems treating Upper Freeport coal. In the case of this coal, t_e was found to correlate well with impeller tip speed for all tank sizes tested (see Figure 40). The equation shown below fits the data well since it has a correlation coefficient of 0.965.

$$t_e = 7140\ V/S^{5.42} \tag{42}$$

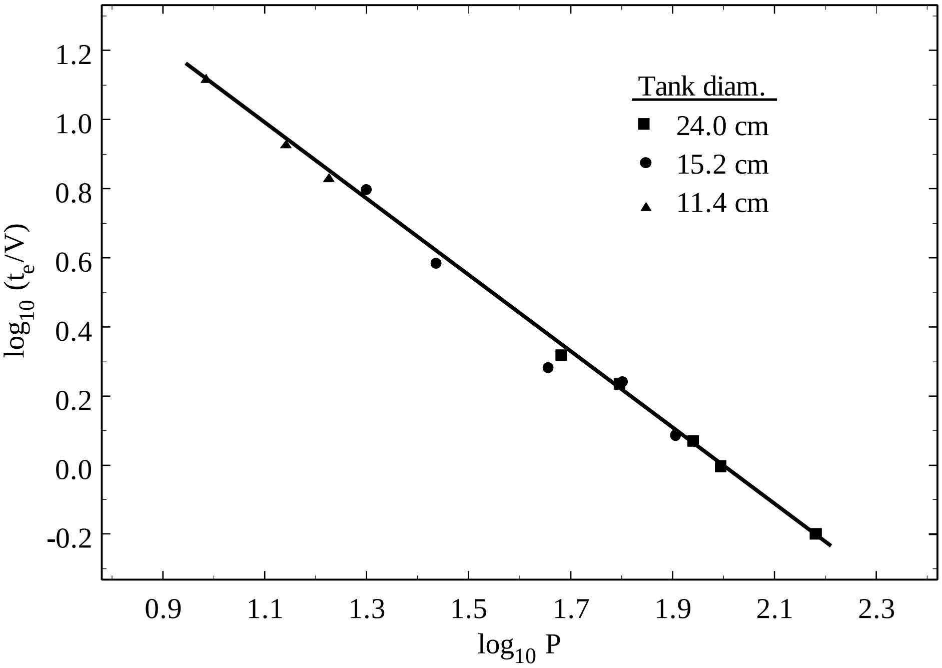


Figure 38. Correlation for t_e observed with Pittsburgh coal and the following conditions: X_1 = 0.30, X_2 = 0.09, X_3 = 0.30, and D/T = 0.3.

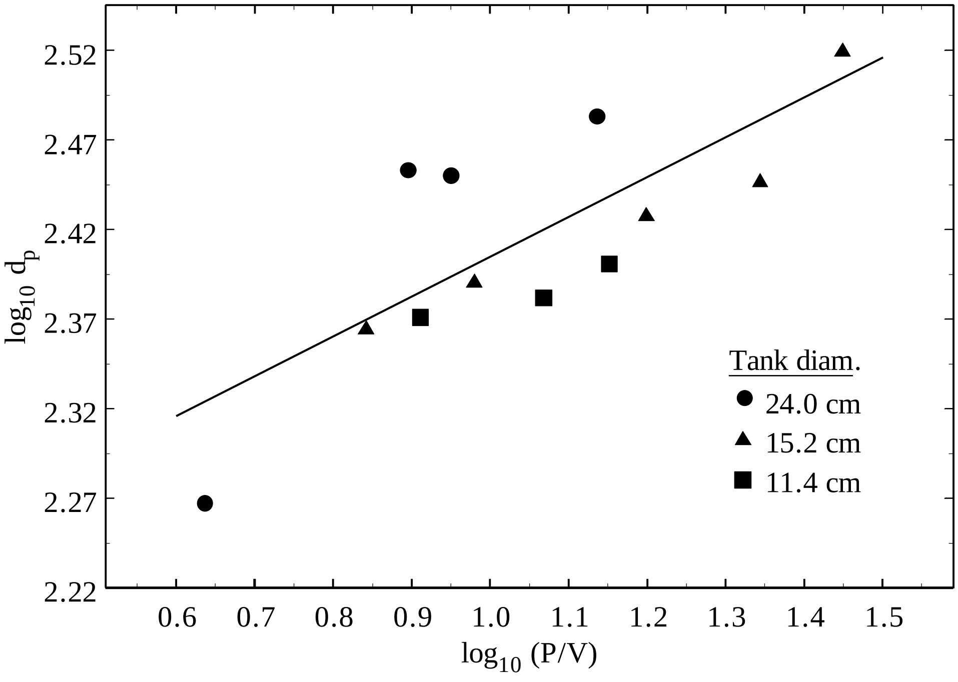


Figure 39. Correlation for d_p observed with Pittsburgh coal and the following conditions: X_1 = 0.30, X_2 = 0.09, X_3 = 0.30, and D/T = 0.3.

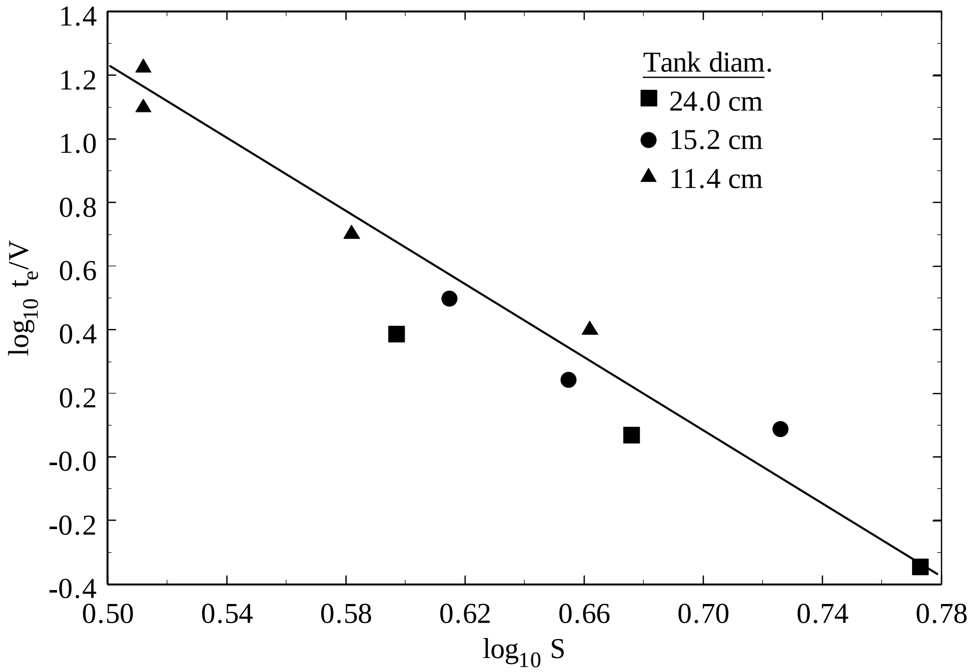


Figure 40. Correlation for t_e observed with Upper Freeport coal and the following conditions: $X_1 = 0.30$, $X_2 = 0.09$, $X_3 = 0.30$, and D/T = 0.3.

Because this equation represents the results obtained with various sizes of mixing systems, it suggests holding $V/S^{5.42}$ constant while scaling up systems for agglomerating Upper Freeport coal.

An alternative basis for scale-up of mixing systems used for agglomerating Upper Freeport coal was not found. The minimum mixing time t_e did not correlate well with agitator power, and d_p did not correlate well with either agitator power or impeller tip speed.

While these results are only tentative because they encompass a relatively small range of mixing tank sizes and limited experimental conditions, they do suggest that there is not a single basis which is universally applicable for scaling up all agglomeration systems. Therefore, the practitioner is still left with the need for conducting agglomeration tests with the intended coal and applicable operating conditions using mixing tanks of various sizes.

CONCLUSIONS

In carrying out the preliminary laboratory-scale development of a gas-promoted oil agglomeration process, the fundamental nature and basic characteristics of the process were established and several tentative conclusions were reached. Most importantly it was shown that gas bubbles have to be present in an agitated system used for agglomerating a moderately hydrophobic coal such as Pittsburgh No. 8 coal. Gas bubbles trigger the process of agglomeration and participate in a very complex mechanism which involves interactions between solid particles, oil droplets, and gas bubbles all suspended in water under very turbulent conditions. The process seems to take place in stages involving dispersion of oil and gas, particle flocculation, coagulation, and agglomerant building. With prolonged agitation, agglomerates are compacted, and given sufficient agglomerant, the agglomerates will combine to form larger framboidal agglomerates. While oil bridges between particles have been regarded as the most likely bonding mechanism for agglomerates, the present work suggests that very small gas bubbles enclosed by water films may serve to hold agglomerates together providing the particles are sufficiently hydrophobic. Moderately hydrophobic coal particles can be made sufficiently hydrophobic by coating them with small amounts of a liquid hydrocarbon such as heptane.

A study of the kinetics of agglomeration of coal particles with liquid hydrocarbons in dilute aqueous suspensions showed that the rate of agglomeration is proportional to the particle number concentration raised to a power between 1.0 and 1.3. The rate increases with increasing amounts of dispersed air or liquid hydrocarbon and with increasing agitator speed. The rate is also greater for Upper Freeport coal than for Pittsburgh No. 8 coal, apparently because of the greater hydrophobicity of the former.

While working with concentrated suspensions of coal particles, the minimum time t_e required to produce compact spherical agglomerates was found to be a key parameter for characterizing the process of agglomeration. Other parameters which were used for characterizing the process of agglomeration included the projected area mean particle diameter d_p of the final agglomerates, the recovery of coal on a dry, ash-free basis, the product ash content, and the amount of ash rejected in the tailings. Since coal recovery and ash rejection were generally large and the product ash content was generally low, these parameters were not as useful as t_e and d_p for characterizing performance.

The results of numerous agglomeration tests with concentrated suspensions of two kinds of coal were correlated empirically for a wide range of experimental conditions. In the case of Pittsburgh No. 8 coal, an excellent correlation was established between t_e and the system volume, agitator power, and the concentrations of solids, i-octane, and air. A good correlation was also established between d_p and i-octane concentration, the ratio of impeller diameter to tank diameter, and agitator power input per unit volume. Although similar correlations for t_e and d_p were established for Upper Freeport coal, they differed in several important aspects. For this coal t_e did not seem to depend on particle concentration, and d_p seemed to depend only on i-octane concentration.

The empirical correlations provide a convenient means for representing and analyzing the characteristics of the agglomeration process within the limits of the variables employed. For example, the correlations showed that t_e is greater for Pittsburgh No. 8 coal than for Upper Freeport coal under similar conditions and showed that t_e decreases more rapidly as agitator speed increases for Pittsburgh coal than for Upper Freeport coal. Consequently, at a sufficiently high agitator speed, t_e is nearly the same for both coals.

For scaling up the size of an agglomeration system it seems appropriate to hold t_e constant. The results for Pittsburgh No. 8 coal suggest that t_e will remain constant provided $V/P^{1.09}$ is held constant during scale up. Of course, this basis only applies for geometrically similar systems and may only apply for the specific set of i-octane, air, and particle concentrations used here. Since the results with Upper Freeport coal indicate a different basis for scale up (i.e., $V/S^{5.43}$ should be held constant), it would be only prudent for any future application of this process to verify the scale up basis. Verification would require conducting further agglomeration tests with mixing systems of different size and using conditions which are specific for the intended application.

Most of the agglomeration tests conducted with concentrated suspensions resulted in a high recovery of a relatively clean product. The recovery was shown to depend on agglomerate diameter d_p which depends on other factors. In addition, the recovery and ash rejection also depend on the procedure used for screening and washing the agglomerates. This part of the process needs further study.

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PUBLICATIONS AND PRESENTATIONS

The publications and presentations resulting from this research are listed below. Additional publications will be forthcoming in the near future:

Publications

F. Zhang, “On the kinetics of the oil agglomeration of coal,” M.S. thesis, Iowa State University, Ames, Iowa, 1995.

J. Drzymala and T. D. Wheelock, “Air agglomeration of hydrophobic particles,” in: *Processing of Hydrophobic Minerals and Fine Coal*, J. S. Laskowski and G. W. Poling (eds.), Canadian Institute of Mining, Metallurgy and Petroleum, Montréal, Canada, 1995, pp. 201-211.

F. Zhang and T. D. Wheelock, “Coal Beneficiation Kinetics of a Gas-Promoted Oil Agglomeration Process,” 13th Annual International Pittsburgh Coal Conference Proceedings, Vol. 1, S.-H. Chiang (ed.), The University of Pittsburgh, Sept. 3-7, 1996, pp. 547-552.

J. Drzymala and T. D. Wheelock, “Preliminary Characterization of a Gas-Promoted Oil Agglomeration Process,” 13th Annual International Pittsburgh Coal Conference Proceedings, Vol. 2, S.-H. Chiang (ed.), The University of Pittsburgh, Sept. 3-7, 1996, pp. 898-903.

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M. Shen, “A gas-promoted oil agglomeration process for cleaning coal,” Ph.D. thesis, Iowa State University, Ames, Iowa, (in preparation).

Presentations

T. D. Wheelock, “Development of a Gas-Promoted Oil Agglomeration Process,” (DOE) University Coal Research Contractors’ Review Conference, Nashville, TN, June 13-14, 1995.

J. Drzymala and T. D. Wheelock (speaker), "Air Agglomeration of Hydrophobic Particles," 34th Annual Conference of Metallurgists, Canadian Institute of Mining, Metallurgy and Petroleum, Vancouver, B.C., August 20-24, 1995.

T. D. Wheelock, J. Drzymala, M. Shen, and F. Zhang, "Development of a Gas-Promoted Oil Agglomeration Process," (DOE) University Coal Research Contractors' Review Conference, Pittsburgh, PA, June 4-5, 1996.

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J. Drzymala and T. D. Wheelock, "Preliminary Characterization of a Gas-Promoted Oil Agglomeration Process," poster presentation, 13th Annual International Pittsburgh Coal Conference, Pittsburgh, PA, Sept. 3-7, 1996.