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# PELLETIZATION OF FINE COALS

(DOE Grant No. DE-FG-22-89PC89766)

# Final Report

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## EXECUTIVE SUMMARY

Coal is one of the most abundant energy resources in the United States with nearly 800 million tons of it being mined annually. Process and environmental demands for low-ash, low-sulfur coals and economic constraints for high productivity are leading the coal industry to use such modern mining methods as longwall mining and such newer coal processing techniques as froth flotation, oil agglomeration, chemical cleaning and synthetic fuel production. All these processes are faced with one common problem area -- fine coals. Dealing effectively with these fine coals during handling, storage, transportation, and/or processing continues to be a challenge facing the industry.

Agglomeration by the unit operation of pelletization consists of tumbling moist fines in drums or discs. Past experimental work and limited commercial practice have shown that pelletization can alleviate the problems associated with fine coals. However, it was recognized that there exists a serious need for delineating the fundamental principles of fine coal pelletization. Accordingly, a research program has been carried involving four specific topics: (i) experimental investigation of coal pelletization kinetics, (ii) understanding the surface principles of coal pelletization, (iii) modeling of coal pelletization processes, and (iv) simulation of fine coal pelletization circuits. This report summarizes the major findings and provides relevant details of the research effort.

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# 1.0 INTRODUCTION

Coal is one of the most abundant resources in the United States with nearly 318 billion tons (including lignite) that can be mined with proven technologies [25]. These reserves will last for several hundred years at the present coal production rate of about 800 million tons per year [25]. Limited availability, increased prices, and dependence on foreign supplies of oil and natural gas have revived interest in coal as a viable source for meeting the national energy requirements. In keeping up with the higher demands for coal and in attempting to keep the raw coal production costs low, the coal industry is using such modern mining methods as longwall mining. The new mining technologies invariably result in non-selective mining and contribute to higher sulfur and ash contents in the raw coals. Subsequent utilization of coal, as an alternative fuel to oil and natural gas, is expected through direct combustion for power generation and through conversion by gasification or liquefaction. If coal is to become our major source of energy, however, it is necessary that the sulfur and ash content of the run-of-mine coals be reduced to meet the stringent EPA regulations. For example, the high-sulfur and high-ash content of the coals fired in utility boilers give rise to the respective environmental problems of acid rains and fly-ash and bottom-ash disposal. Accordingly, such advanced coal cleaning techniques as froth flotation and oil agglomeration are being applied to reduce the sulfur and ash contents of raw coals.

Both the newer mining and coal cleaning technologies contribute to the generation of large quantities of coal fines. For example, according to one estimate, the production of -28 mesh fines has increased from 14% to 31% (by wt.) due to mechanized mining [25]. An even larger

portion of fines is generated at the coal cleaning plants where finer grinding of coal is required to liberate and remove ash and sulfur from combustible matter.

## 1.1 Fine Coals - Problems and Solutions

There are several problems in dealing with fine coals and the more important are listed in the following.

- The use of conventional and newer techniques to clean the finely crushed and ground coals leads to inefficiency and loss of significant amounts of fines into the refuse streams of coal preparation plants. It is estimated that there is a billion tons of coal potentially recoverable from the refuse ponds in the U. S. [42].

- Dewatering of fine coals is quite inefficient giving rise to higher moisture contents of the filter cake and often necessitating the additional costly step of thermal drying. Thermal drying of coal fines leads to such problems as spontaneous combustion and particulate pollution. To avoid these, proper control of the dryer and expensive dust abatement equipment become necessary.

- During transportation of fine coals from preparation plants to power plants, significant amounts of coal fines are lost as dust. According to one study, losses as high as 3% can occur [46]. Besides the obvious loss of combustible matter, the lost material constitutes serious environment pollution.

- Finally, during storage and handling there are the perennial problems, for both the coal industry and the consumer, of spontaneous combustion, dust pollution and freezing (during the winter season).

Agglomeration, a method of forming larger bodies from fine particles [31, 56, 62], is capable of alleviating most of the problems encountered with fine coals. The method is successfully used in mineral, metallurgical, fertilizer, and various other process industries. There are several techniques available for agglomeration of fine particles and these can be conveniently classified into three categories.

- agglomeration of liquid-suspended solids which include flocculation, selective flocculation and oil-agglomeration;

- agglomeration by pressure compaction methods including briquetting and extrusion; and

- agglomeration in rotating-beds or fluidized-beds which include pelletization, balling, granulation.

In the coal industry, one of the above methods of agglomeration could be readily selected depending on the specific problem that is being addressed.

Flocculation is found to be effective in improving the solid-liquid separation (thickening and filtration) and oil agglomeration is close to commercial application for the recovery and dewatering of fine coals.

Briquetting is the oldest agglomeration technique employed in the coal industry and is continued to be used. However, it is considered most suitable for dry coals and small tonnages. Extrusion is an established process in the ceramic and food industries and works efficiently on feed materials that are plastic in nature; for coal, this requirement is normally met by addition of plasticizers or heat treatment. A recent investigation has shown that a roller-die type extrusion can be most economical in an agglomeration/reslurrying scheme as a means of distributing and firing clean coal [10].

Agglomeration of coal fines in fluidized-beds is expected neither to be feasible nor economical. On the other hand, pelletization is a well established technology and involves tumbling of moist fines in rotating drums or discs. The pelletization process is more commonly known as balling in the iron ore industry and granulation in the agro-chemical industry. This process was commercially applied to coal fines and has the potential of being fully integrated in coal preparation plant flowsheets.

## 1.2 Project Objective and Specific Research Topics

Published literature on pelletization of coal fines is somewhat limited. Though the first paper concerning the pelletization of coal fines appeared in 1955, since then no more than fifty papers

have been published. Most of these papers are applied in nature and only a few of them addresses the principles of coal pelletization (see Section 2).

The primary objective of this undertaking was able to generate fundamental information that would be of practical significance to the coal industry. Specifically, the research was directed to four main topics:

1. Experimental investigation of coal pelletization kinetics,
2. Understanding the surface principles of coal pelletization,
3. Modeling of coal pelletization processes, and
4. Simulation of fine coal pelletization circuits.

## 1.3 Organization of the Final Report

This final report summarizes the major findings and provides relevant details of this research effort. It is organized into 8 sections. The first section provides a general introduction to the report. The second section provides a literature survey on coal pelletization and the details about the materials and experimental methods are included in the third section. The next two sections (Sections 4 and 5) present respectively an account of the experimental results and an analysis of the results leading to the development of suitable mathematical models for coal pelletization. Then, Section 6 introduces *CoalAgglom*, an interactive simulator for studying the dynamic behavior of fine coal pelletizing circuits. Finally the report is concluded with Sections 7 and 8 in which Summary and Conclusions and List of References are included.

# 2.0 BACKGROUND

In the following we present a general overview of the pelletization process and then highlight specific aspects of coal pelletization.

## 2.1 General Aspects of the Pelletization Process

Pelletization is the process of forming nearly spherical pellets by tumbling moist particulate fines (with or without addition of binders and other necessary additives) in such devices as drums, discs, or cones. Mineral, ceramic, chemical, food, fertilizer, nuclear, pharmaceutical, waste processing and a host of other industries make use of pelletization to alleviate problems associated with fine particles. However, from a tonnage stand point, pelletization is most commonly used for agglomeration of iron ore concentrates. Detailed reviews of various practical and theoretical aspects of the pelletization of fine particles have been published [21, 32, 56].

As shown in Figure 2.1, the driving potential for the pelletization of fine particles is the lowering of the total surface free energy of the system through a reduction of the effective air-water interfacial area [56]. The forces which bring about this change are of two kinds: (a) physical and (b) applied [55]. The physical forces responsible for the formation and the strength of pellets arise from a number of sources - (i) interfacial effects giving rise to capillary and surface tension forces, (ii) van der Waal's interaction, magnetic and electrostatic interactions, (iv) interlocking effects among the particles, and (v) immovable bridging bonds []. The applied forces which arise due to the tumbling motion of drums or discs serve the purpose of bringing individual liquid-wetted particles into proximity with each other so that the physical forces become operative and pelletization is initiated. Calculations by Rumpf [50] indicate that, for the

$$\Delta G_{agglom} = (S_{nuclei} - S_{feed})\, \gamma_{air-water}$$

Figure 2.1. A schematic showing the driving force for pelletization due to reduction in total air-water interfacial area.

case of solid particles which are completely wetted by a bridging liquid, capillary and surface tension forces are the most predominant. The magnitude of capillary forces depends upon the type of bond between the particles. These bonds, depending upon the relative proportion and distribution of the air and water phases in the pellet are referred to as (i) pendular, (ii) funicular, and (iii) capillary bonds.

The changes in the state of the agglomerating system can in principle be accounted in terms of the above described forces, but it is simpler to make use of the concept of size change mechanisms. Such a view point is advantageous because of the inadequate understanding of the occurrence of these various forces at a molecular or microscopic level. A total of seven mechanisms of agglomerate formation and size change was suggested earlier [55]. These include (i) nucleation, (ii) dissociation, (iii) coalescence, (vi) breakage, (v) layering, (vi) attrition, and (vii) abrasion transfer. For the purpose of our discussion it is enough to consider three of the seven mechanisms - nucleation, coalescence, and layering (see Figure 2.2).

Nucleation refers to the formation of particles from the continuous phase due to interactions within the environment in itself. Clearly, nucleation causes appearance of new particles along with a discrete change in the size of the newly born agglomerates. As soon as the initial formation of nuclei occur, coalescence begins to set in. It causes discrete size changes and leads to decrease in the number of agglomerates but not their mass. The third mechanism of layering corresponds to continuous size change of the particles and is affected by interaction of particulates with the environment (continuous phase). Accordingly, layering brings changes in the mass of the agglomerates but not their number. Clearly, layering can occur only when both the agglomerates and new feed are present.

MOIST FEED, $p_i$    PELLETS (well-formed species), $P_i$

NUCLEATION

$$j\,p_i \rightarrow P_j$$

COALESCENCE

$$P_i + P_j \rightarrow P_{i+j}$$

LAYERING

$$P_i + j\,p_i \rightarrow P_{i+j}$$
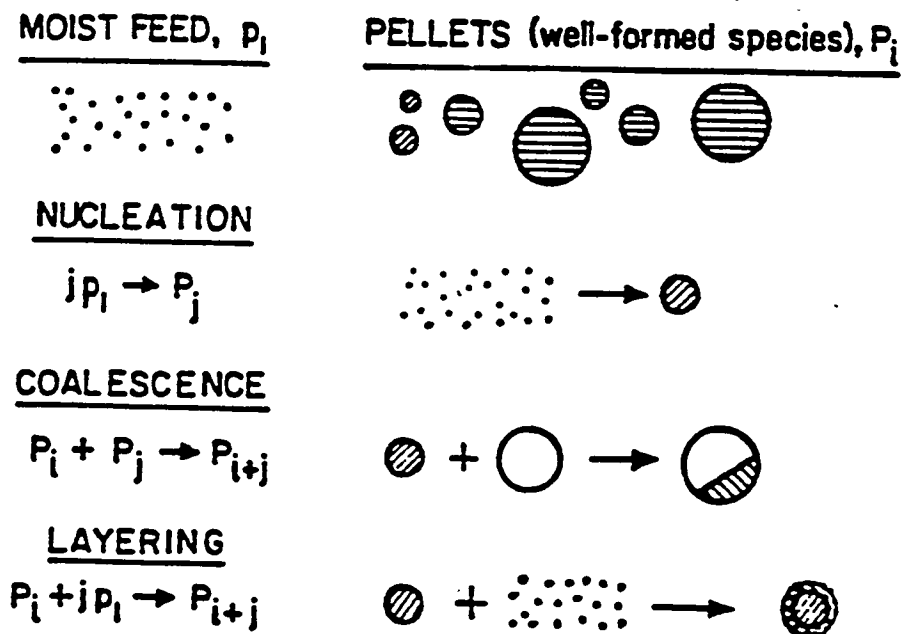
Figure 2.2.  Three main mechanisms of pellet formation and growth: nucleation, coalescence, and layering.

These mechanisms take place simultaneously during any agglomeration process unless the design and operating conditions are otherwise chosen. Actually, the simultaneous occurrence of these mechanisms is known to result in undesirable agglomerate size distributions and cycling of the performance of an agglomerating unit. Also it is recognized that different growth mechanisms can prevail depending upon the type of feed being pelletized. According to Kapur and Fuerstenau [31], the growth of pellets in the initial stages of batch pelletization proceeds first by nucleation and then mainly by the coalescence mechanism. In later stages of pelletization there is evidence that a limited amount of breakage of pellets occurs. In industrial pelletizing operations where a continuous supply of moist feed is charged to the pelletizing device, the growth mechanism of layering (or snowballing) of added moist feed onto the pellets becomes important.

In the past, the following types of laboratory experimental techniques have been used to study the pelletization behavior of particulate materials:

1. The first technique consists of slowly adding moist feed together with spray water onto a small batch of seed pellets inside a rotating airplane tire to produce large pellets. A detailed account of this technique, which is popular in iron ore processing laboratories for studying the strength and induration characteristics of pellets, is given elsewhere [66].

2. The second technique is a batch experimental method that consists of tumbling a charge of moist feed in a laboratory drum without further addition of water and/or moist feed [54, 57]. This technique is most suitable for studying pellet growth mechanisms, pellet growth kinetics, influence of process variables, and role of additives.

3. The third technique known as the locked-cycle pelletization technique is a semi-continuous experimental procedure that is used to simulate the closed-circuit drum operation in a laboratory [53].

Using the batch experimental technique, it has been established by Kapur and Fuerstenau [31] that the batch pelletization kinetics of comminuted feed materials is typically described by an S-shaped curve. This S-shaped curve shows three distinct regions of growth (depending upon

the operating conditions) termed as - (i) the nuclei growth, (ii) a transition growth, and (iii) the pellet or ball growth regions. The extent of these growth regions and hence the kinetics of pellet growth depends strongly on (i) the moisture content of the feed, (ii) the size distribution and fineness of the feed, (iii) the feed preparation method, and (iv) the addition of binders. The size distribution of the resulting pellets in a batch experiment has a self-preserving nature which implies that irrespective of the operating conditions and the feed material, the size distribution, when normalized with respect to the average size of the distribution, is represented by a single curve. This characteristic size distribution is called the similarity size distribution. Sastry [52], using a population balance approach developed mathematical models and computer simulation of the pellet growth in balling drums and discs.

Strength characteristics of wet and dry pellets has been a subject of a few theoretical and several experimental investigations. A detailed review of the literature on the strength of wet pellets has been prepared by Kapur [32]. It is now fairly well established that the strength of moist pellets is mainly due to capillary forces. However, in the presence of particles of colloidal dimensions, the capillary theory alone is not adequate to account for the wet strength. Also, the capillary forces, which are important in moist pellets disappear once the pellets have dried and then the forces contributing towards the dry strength of a pellet arise from (i) hardening of bonding agents, (ii) precipitation of dissolved salts, (iii) friction and interlocking between particles , and (iv) electro-static, magnetic or van der Waal's interactions. In order to improve the strength characteristics of the pellets, binders have been commonly used in the iron ore and other industries. Bentonite, which is the most common additive for iron ore pellets, has been extensively studied as a binder. A large number of other additives have been investigated as possible substitutes for bentonite [61], with demonstrable commercial success.

## 2.2 Pelletization of Coal Fines

The first review paper on coal pelletization was published as early as 1962 [4] and since then about fifty papers have appeared in the literature. More recent literature reviews can be found in [7, 36, 38, 44, 57, 59, and 62]. Following is a brief overview of selected papers from the literature covering the laboratory investigations and industrial practice of coal pelletization.

### 2.2.1 Laboratory Studies

The first report on coal pelletization was published by Carveth and Lang [7] in 1951. This study examined the results of blending coal fines with hot asphalt and steam while maintaining the mix at 100° C. This process generated pellets ranging from 35 to 50 mm diameter. However, the process itself was not considered commercially viable due to high heating costs and high asphalt requirements.

Handfield-Jones [23] investigated the pelletization of froth floated coals. That study observed that the strength of coal pellets could be increased by adding bentonite or fine shale as binders. The study linked the growth behavior of coal pellets with the distribution of water within the pellet. Also, it was found that the strength of the pellet was affected by the colloidal content of the binders.

Sastry and coworkers [36, 59] carried out an extensive study on the pelletizability of coal fines from 12 bituminous coals, 2 subbituminous coals, 1 anthracite and 1 lignite. This study focused on the effect of feed size, feed moisture, and binder addition on process kinetics. They concluded that coal fines could be pelletized with a starch binder and that the economics of the coal pelletizing process is primarily determined by the cost of binders and the cost of thermal drying of the pellets.

Pelletization of coal fines is found to be possible only within a narrow range of moisture additions; outside the range, the coal fines are either too dry or too wet to agglomerate properly [36]. Ash content of coal is the most significant property determining the moisture requirements for pelletization. For example, coals with 5% ash require 43% (by weight) moisture, whereas those with 20% ash require only 30% water addition. The moisture requirement for pelletization, W (expressed as weight percent of dry coal), and the percent ash in the coal fines, A, have an empirical relationship:

$$W = 43.96 \exp(-0.017\ A).$$

This moisture-to-ash relationship holds for bituminous and anthracite coals and not for lignite. Lignite requires much higher moisture for pelletization than estimated from the above relationship. This is explained in terms of the internal pore structure of lignite. Further, the sulfur content of coal is found not to influence the pellet growth behavior. The surface area of coal fines is an important variable influencing the kinetics of pellet growth, in that, for anthracite and bituminous coals, the rate of growth decreases with increasing fineness of feed, whereas for lignites a reverse trend is observed.

Among other binders, bentonite, asphalt emulsion, and starch are the three binders most commonly investigated and used during coal pelletization [36, 68, 69]. Of the three binders, corn starch is found to be most effective in improving the wet and dry pellet compressive strengths and abrasion and impact resistances but not resistance to water penetration. Corn starch binder is effective for the pelletization of anthracite, bituminous as well as lignite type coal fines. An addition of 0.5% (by wt.) appeared to yield pellets with adequate quality. Bentonite, which is popular in the iron ore industry, is not found to perform as well during coal pelletization [36].

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Asphalt emulsion is found effective in providing excellent protection against water penetration. This ability of asphalt emulsion has been attributed to its hydrophobic surface nature. A combination of corn starch (imparting strength) and asphalt emulsion (imparting resistance to water penetration) was found to yield the best quality coal pellets.

Rubio and De Leon [49] compared the pelletizing and briquetting results of Brazilian coal fines. They detailed results from varying moisture content, binder type and concentration, and curing conditions. Their conclusion was that pelletizing was more economically favorable than briquetting due to the extra drying costs in the briquetting process.

Most recent studies by Wen, et.al. [68] have shown that a humic acid binder, consisting of a water soluble colloidal system of humic acids dispersed in ammonium hydroxide solution, produces strong and durable bituminous coal pellets [31, 32].

Onyemaobi [47] used a set of froth-floated British coals to show that hydrophobicity exerts more of an effect on moisture requirements for pelletizing than ash content.

A study by Conkle, et. al [10] was directed to investigate the pelletizing and reslurrying as a means of distributing and firing clean coal. Laboratory agglomeration studies were carried out with disk pelletizer, extrusion and roller-die equipment. It was found that all the three methods yield satisfactory coal agglomerates for reslurrying, but roller-die method is expected to be most economical.

While there are very few studies on the role of surface properties on coal pelletization, there is an abundance of publications in the context of coal cleaning. For example, a recent publication by Aplan [1] includes a review of the influence of hydrophobicity on coal preparation operations such as flotation, filtration, flocculation, dispersion, storage and handling.

## 2.2.2 Industrial Practice of Coal Pelletization

Coal pelletization was commercially implemented for the first time by the Rochester and Pittsburgh Coal Company [7, 9]. Feed for this operation originated from coal fines recovered from waste slurries by froth flotation. The fines (100% minus 28 mesh, 50% minus 325 mesh) were pelletized in a balling drum after 26% moisture and 0.5 to 1.0% lignin was added. The product pellets (6-20 mm) were dried at 105°C. The operation was considered successful and produced pellets at a rate of 15 tph. The pelletizing process continued for several years until the mine supplying the fines closed.

The Eastern Coal Company [14] also used a froth flotation process to recover coal fines from a waste stream as feed to a disc pelletizer. This process used 1.5 weight percent lignin liquor as binder. While the pelletizing step was reported as successful, serious problems occurred during their thermal drying, in that the pellets caught fire when their moisture fell below 1%. The plant was later shut down because of this problem.

Hinkle [24] reported on producing agglomerates from coal fines recovered from a power plant dust recovery system. Several methods of generating acceptable agglomerates were studied. The first test method involved combining medium sized lumps of coal with dust. However, this method produced excessive fines when the final agglomerate product was being returned to the coal delivery system. The final method that was developed to produce usable pellets with minimal dust generation involved an Ampel mixer. This report is one of the few successful implementations of industrial scale coal pelletization.

Conceptual design studies indicate that a typical coal pelletizing plant would consist of a pelletizing drum or disc circuit with a roller screen and a roller dryer. Capital cost of a 100 TPH pelletizing plant (including dryer) appears to be in the neighborhood of $15 million. The

operating costs are estimated to be $12 per ton of coal pelletized out of which binder and drying costs represent 60% [57].

Jones and Meecham [29, 30] present some of the latest efforts to utilize coal pelletization in a full-scale application. They describe efforts at British Coal to solve problems of coal fines handleability. British Coal installed and tested a pilot plant, but did not progress to full production.

## 2.3 Conclusions

We may conclude from the above literature survey that research activity in coal pelletization is quite limited and most of it is of applied type. There exists a clear need to understand the process engineering aspects of the coal pelletization process including studies on the influence of operating variables, role of surface chemical properties of coal and quantitative modeling and process simulation.

# 3.0 MATERIALS AND METHODS

## 3.1 Coal Samples

The pelletization experiments were carried out on three bituminous and one anthracite coal samples. These samples were provided courtesy of a concurrent U.S. Department of Energy contract studying "Coal Surface Control for Advanced Fine Coal Flotation" (DOE Project No. DE-AC-88PC88878) [18]. We have chosen these coals for two specific reasons - (i) they are extensively characterized for physical, chemical, petrographic and surface properties and flotation behavior and (ii) they are readily available in Berkeley. The list of coals and mine locations from which they were sampled is included in Table 3.1.

Details of the coal sample collection were given by Fuerstenau et.al [18]. Here we include only the highlights of the sampling program. Extensive care was used in the collection and handling of the run-of-the-mine samples. Fresh coal samples were collected from new mine faces after the removal of clay and rocks. Topsize material was then reduced to minus 4 inches in the field, homogenized and loaded in 55-gallon drums. Approximately 400 pounds of coal were loaded per drum. Each drum was then repeatedly filled with argon gas and sealed ("inerted") to minimize oxidation.

## 3.2 Preparation and Characterization of Fine Coal Samples

Coal fines were produced under controlled conditions by stage crushing and grinding run-of-mine samples. The procedure consisted of first stage of crushing a 10 kg sample of the 4 inch as-received coal down to a nominal 3/4 inch topsize in a laboratory jaw crusher. The second stage consisted of reducing the 3/4 inch feed to a nominal -1/4 inch topsize in a laboratory roll crusher. This 1/4 inch product was then split into 500 gram samples and stored in air-tight bags

Table 3.1. List of coal samples and their sources.

| Seam | Mine Operator | Mine Location |
|---|---|---|
| Bethlehem Anthracite | Bethlehem Mines Co. | Tamaqua, PA |
| Illinois No. 6 | Peabody Coal Co. | Marissa, IL |
| Pittsburgh No. 8 | R & F Coal Co. | Warnock, OH |
| Upper Freeport PA | N. Cambria Fuel Co. | Blacklick, PA |

under "inert" conditions. Each 500 g sample of stage crushed coal was first dried in a forced air heater at 50°C for about 12 hours and was dry ground for 30 minutes in a 8 inch diameter by 10 inch long stainless steel laboratory mill (loaded with 16, 14, and 12 stainless steel rods of 3/4, 5/8, and 1/2 inch respectively). All the pelletizing experiments were conducted within hours of producing the fine coals by dry grinding. This procedure was developed to minimize the possible oxidation of the fines.

All the fine coal samples were characterized for their size distribution, specific surface area, proximate and ultimate analysis. Following sections present the details of the test procedures.

### 3.2.1 Particle Size Distribution

The standard wet and dry sieving technique was used for determining the particle size distribution of fine coal samples. It consisted of wet sieving 100 g of coal sample on a 400 mesh screen and drying the +400 mesh fraction and subjecting it to dry sieving on a nest of sieves. Table 3.2 presents size distribution of the dry ground coal feeds used for the batch pelletization experiments.

### 3.2.2 BET Area

Specific surface area of the fine coal samples was determined by gas adsorption using a Micromeritics BET Surface Area and Pore Size Analyzer Model 2100. The adsorbate gas used was a research grade carbon dioxide (purity 99.998%). The surface area was calculated using the Dubinin-Polyanyi equation and adsorption data. The results which are included in Table 3.2 show that Bethlehem anthracite has the highest surface area, while the three bituminous coals, Illinois No. 6, Pittsburgh No. 8, and Upper Freeport PA having similar surface areas of roughly 160 to 180 $m^2$/gm. Fuerstenau et al. [18] found that changing the grind conditions of these coals did not significantly alter the surface area of the coals. The apparent lack of change in surface

Table 3.2 Physical and chemical characteristics of coal samples used in this project.

| | Bethlehem Anthracite | Illinois No. 6 | Pittsburgh No. 8 | Upper Freeport PA |
|---|---|---|---|---|
| **Size Distribution, cumulative weight percent passing** | | | | |
| Mesh (opening in $\mu$m) | | | | |
| 28 (600) | 100 | 100 | 100 | 100 |
| 48 (300) | 95 | 100 | 100 | 99 |
| 65 (212) | 96 | 99 | 94 | 97 |
| 100 (150) | 77 | 99 | 82 | 89 |
| 150 (106) | 57 | 97 | 65 | 72 |
| 200 (75) | 36 | 83 | 48 | 54 |
| 325 (45) | 23 | 57 | 32 | 36 |
| 400 (38) | 10 | 30 | 12 | 8 |
| | | | | |
| **Surface Area, $m^2$/g** | 380 | 165 | 150 | 150 |
| | | | | |
| **Proximate Analysis, weight percent -- dry basis (*)** | | | | |
| Fix. Carbon | 92.9 | 46.3 | 43.5 | 61.8 |
| Ash | 3.5 | 16.4 | 11.5 | 13 |
| Volatiles | 3.7 | 36.2 | 35.6 | 26.2 |
| Total Sulfur | <0.1 | 5.73 | 4.89 | 2.38 |
| As Received | | | | |
| Moisture | 2.4 | 9.5 | 2.3 | 1.0 |
| | | | | |
| **Ultimate Analysis, weight percent -- dry basis (*)** | | | | |
| Moisture | 2.4 | 9.5 | 2.3 | 1.0 |
| Carbon | 89.9 | 63.8 | 71 | 75.6 |
| Hydrogen | 2.3 | 5.7 | 5.1 | 4.7 |
| Nitrogen | 1.16 | 1.24 | 1.45 | 1.45 |
| Oxygen | 1.18 | 6.1 | 6.4 | 3.85 |
| C/O Ratio | 76.2 | 10.5 | 11.1 | 19.6 |

(*) Source: Fuerstenau, et.al.[18]

area is typical of high-surface area materials since the change in external surface area is small compared with the relatively large surface area in the pores.

### 3.2.3 Proximate and Ultimate Analysis

The proximate analysis was carried out using a LECO MAC 400 Proximate Analyzer. The moisture, volatile matter, ash and fixed carbon were determined gravimetrically by heating the samples at the respective temperatures of 106°C, 950°C, and 750°C. The MAC 400 maintains a nitrogen atmosphere in the furnace during the moisture and volatile analyses, and then uses a oxygen atmosphere in the ash analysis. Sulfur analysis was performed with a LECO SC-132 Sulfur Analyzer. Samples are combusted in an oxygen atmosphere where the sulfur oxidizes to sulfur dioxide. The combustion gases are then analyzed by a solid state infrared absorption detector. Proximate analysis results are presented in Table 3.2.

The ultimate analysis of the three coals was determined by using a LECO CHN 600 Analyzer. The carbon, hydrogen, and nitrogen contents of each coal were obtained by burning samples in pure oxygen at 950°C in an infrared analyzer which measures the amount of $CO_2$ and water vapor in the combustion gases. The amount of nitrogen is determined by a thermal conductivity cell which first converts nitrous oxide to nitrogen gas by passing the hot combustion gas over hot copper and a catalyst. The ultimate analysis of each of the coals is also included Table 3.2.

### 3.3 Additives for Coal Pelletization

During the course of this project, we investigated the influence of two surface active agents that are commonly used in fine coal processing, viz., a flotation collector and a flotation depressant.

### 3.3.1 Dodecane - a Flotation Collector

In coal flotation, a combination of collector and frothers is generally used. Coal flotation collectors are usually nonpolar oils obtained from petroleum distillation or coke-oven plants and therefore have low solubility in water. Typical nonpolar collectors are kerosene, fuel oil, crude petroleum and coal tar distillates. By far, kerosene is the most widely used collector in coal flotation. It is a mixture of saturated paraffinic hydrocarbons whose exact composition depends on the source of the crude and the distillation process and operation. To ensure a consistent quality of collector, dodecane was chosen as a standard collector, since its molecular weight is close to kerosene. Reagent grade n-dodecane obtained from Eastman Kodak Company was used in this study.

### 3.3.2 Dextrin - A Flotation Depressant

Dextrin has been extensively studied as a depressant in various flotation schemes with iron ore by Iwasaki and Lai [27] and with coal Miller et al. [43]. Dextrin is the product of the hydrolysis of starch by certain enzymes. The hydrolysis reaction produces a modified starch that is hydrophilic with a high molecular weight. Dextrin is a short chain molecule that is not likely to act as a binder. The flotation studies indicate that dextrin adsorbs with hydrophobic bonding and that adsorption density increases with an increasing number of hydroxyl groups. The dextrin used for this study is reagent-grade dextrin obtained from VWR Scientific Company.

### 3.4 Batch Pelletization Experimental Procedure

All the batch pelletization experiments were carried out in a 150-mm length by 225-mm diameter pelletizing drum (Figure 3.1) by making use of the procedure that was extensively to investigate the pelletization behavior of different kinds of fine powders [31, 36, 57, 60].
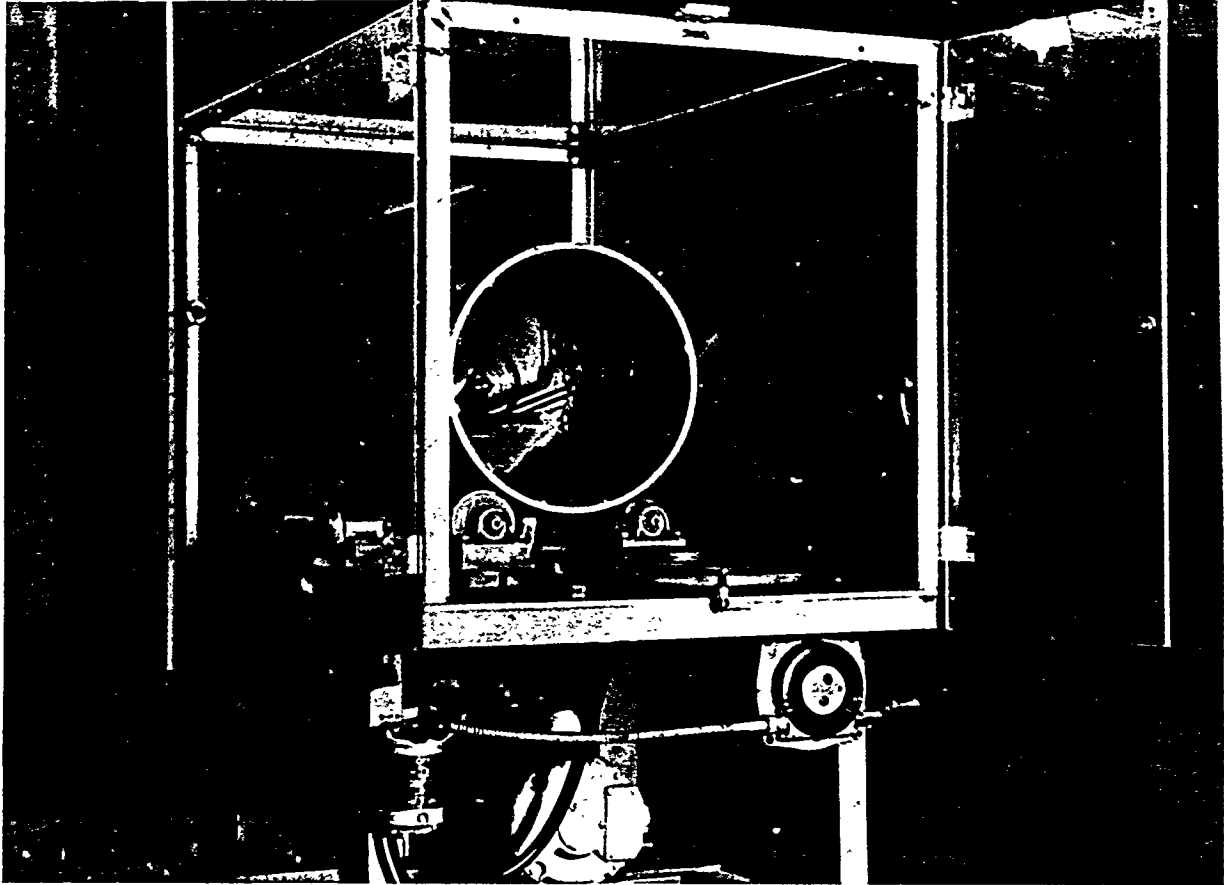
Figure 3.1. Photograph of the batch balling drum assembly.

The first step of a batch pelletization experiment is the preparation of moist feed. It consists of -- (i) placing 250 g of dry fine coal feed in a resealable plastic bag and adding the desired quantities of dextrin/water or dodecane/water emulsion and (ii) sealing the bag and kneading the contents thoroughly by hand. After the contents are thoroughly mixed, the moist coal fines are rubbed through a 6-mesh screen to prepare a fluffy feed that is lump free.

The moist fine coal feed is then loaded into the batch pelletizing drum which is enclosed in a humidified chamber (Figure 3.1) and maintained at 95% relative humidity to minimize the moisture loss from the drum contents. The drum is rotated at a speed of 40 revolutions per minute and batch pelletization is carried out for the desired number of revolutions. At the end of the run, the pelletizing drum is emptied and the drum charge is analyzed for the moisture content and size distribution of the pellets. Each run is repeated 2 to 4 times under identical conditions to establish reproducibility. During each run, small samples of moist feed and final pellet product are taken and their moisture content determined.

Each kinetic experiment then consisted of determining the pellet size distributions as a function of time (drum revolutions) under given operating conditions.

The pellet size distributions were was determined by placing the green pellets on a set of sieves of 1/4 inch to No. 20 Tyler mesh and gently sieving the pellet charge. In the past, the pellet size distributions were determined by photographing and image analysis [32, 57] but we have found that the sieve analysis method gave results much faster. From the sieve analysis we obtain the mass of pellets ($m_i$) retained on the $i$-th sieve and (with a sieve opening of $d_i$) and passing the $(i-1)$th sieve. Then for each pellet sample, we calculate mass fractions ($w_i$),

cumulative mass fractions ($W_i$), average pellet diameter ($\bar{D}_w$) and standard deviation ($\sigma_w^2$) by using the following expressions.

$$w_i = \frac{m_i}{\sum m_i} \tag{3-1}$$

$$W_i = \sum_{k=1}^{i} w_k \tag{3-2}$$

$$\bar{D}_w = \sum \bar{d}_i w_i \tag{3-3}$$

$$\sigma_w^2 = \sum (\bar{d}_i - \bar{D}_w)^2 w_i \tag{3-4}$$

where

$$\bar{d}_i = \sqrt{(d_i d_{i-1})} \tag{3-5}$$

the geometric mean diameter of the agglomerates in the $i$-th sieve fraction.

# 4.0 EXPERIMENTAL RESULTS AND DISCUSSION

In this section we present results from various pelletization experiments that were performed during the course of the project along with a qualitative discussion of the results. The next section deals with the quantitative analysis of the coal pellet growth processes.

## 4.1 Coal Pellet Size Distributions

As mentioned in the last section, each batch pelletization experiment consists of placing a moist fine coal sample in the batch balling drum and tumbling the drum for a predetermined time period. Figure 4.1 shows a picture of -- (a) dry fine coal sample, (b) moist feed just before it is placed in the drum and (c) the spherical pellets that are formed after 800 drum revolutions.

It is instructive to observe that while dry coal fines are extremely dusty, the moist feed to the drum shows a definite degree of prenucleation (with agglomerates in the size range of 0.5 to 2 mm) and has as such less of a dusting problem. Nevertheless the drum feed tends to be fairly fluffy. Tumbling the moist fines for 800 revolutions give rise to near spherical pellets in the size range of 5 mm to 15 mm.

Figure 4.2 shows the progression of pelletization where the moist feed and samples of coal agglomerates at 100, 200, and 400 revolution are shown. What is quite fascinating is that the fluffy feed transforms into well-formed nuclei within 50 or so drum revolutions. During this initial few revolutions the moisture distributes around the particles and the pendular forces pull the individual particles closer giving rise to stable nuclei. These nuclei for the most part tend to grow by coalescence while forming into a larger cohesive spherical shaped pellets with little identity of the individual nuclei.

(a)

(b)

(c)
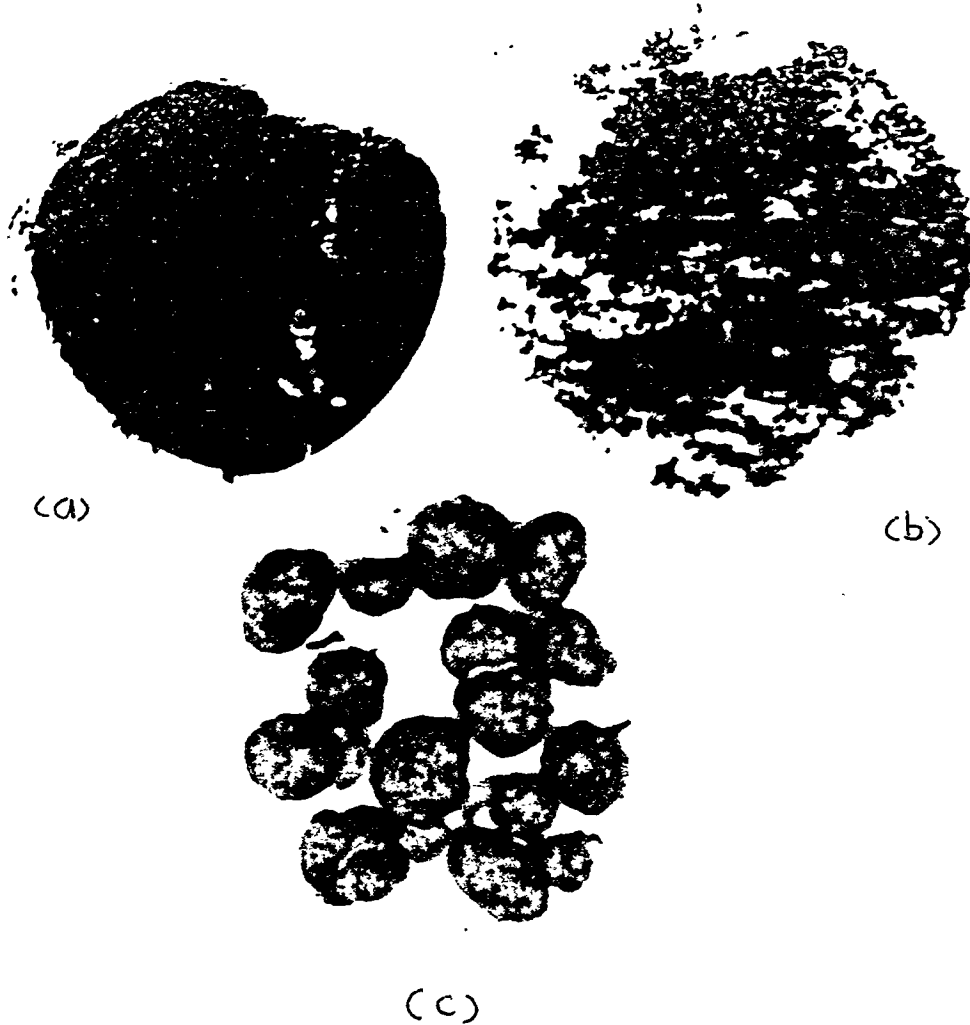
Figure 4.1. Photograph of the Upper Freeport PA -- (a) fine coal sample, (b) moist feed to the balling drum and (c) agglomerates produced after 800 drum revolutions.
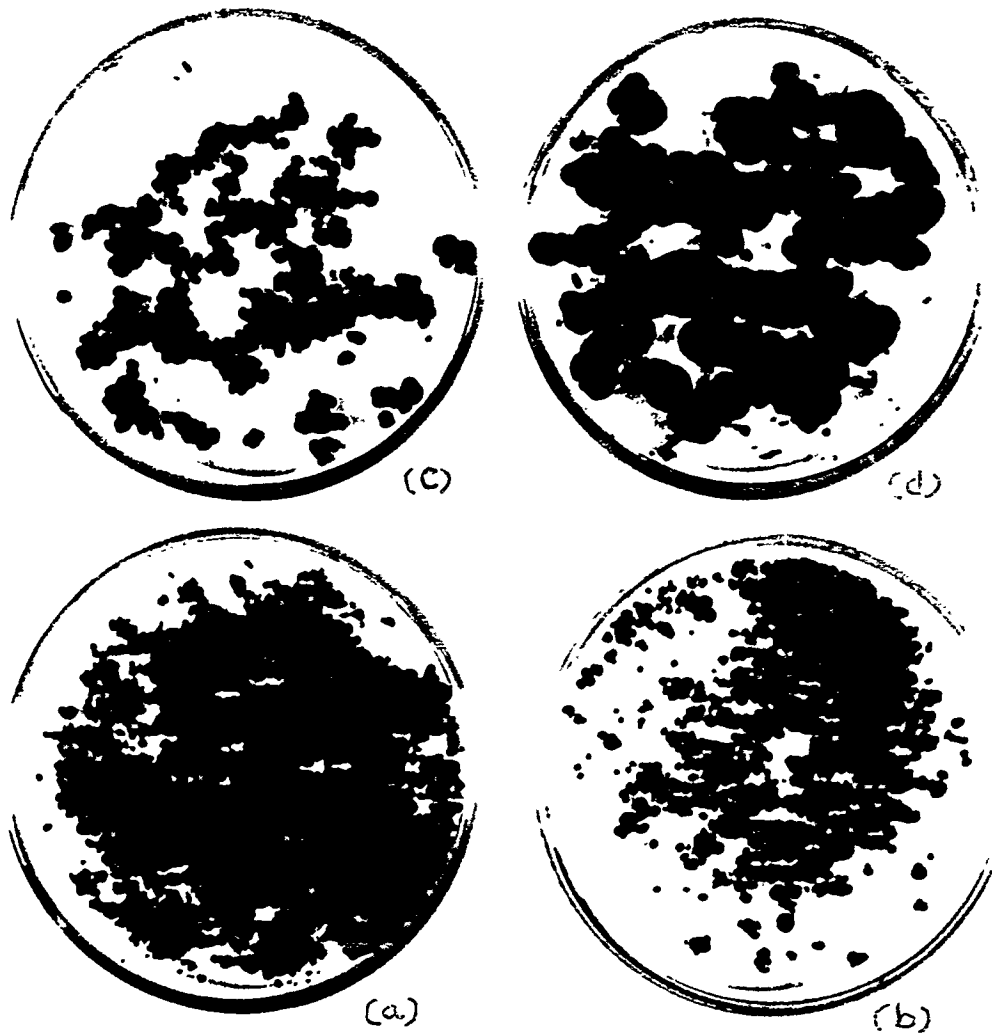
Figure 4.2. Photographs of Upper Freeport PA -- (a) moist feed to the balling drum and agglomerates generated after (b) 100, (c) 200 and (d) 400 drum revolutions.

On the other hand, a few samples tend to grow rapidly by coalescence into raspberry shaped pellets where the individual granules that came together do not deform and keep their identity. We have observed this to be a problem more so with extremely fine coal feed samples. Figure 4.3 shows the spherical and raspberry type coal pellets that we have encountered during our experimental studies. Needless to say the raspberry growth is highly undesirable because the agglomerates tend to be rather porous and weak. Figure 4.3 also includes a mechanistic representation of the formation of spherical agglomerates with binary coalescence and cohesive growth as well as the raspberry type growth.

Figure 4.4 presents the size distribution of agglomerates at various stages of their growth. It is seen that the size distribution gets broader as the agglomerates grow larger. Depending on the intended use of the coal pellets such as dust free transportation, such widening of the distribution might be desirable because it leads to higher packing density of all the pellets put together. It is often reported that most of the agglomerate size distribution obtained from batch pelletization experiments conform to a similarity size distribution. We shall quantitatively analyze the size distribution results from this study in the next section.

From a practical stand point analyzing pellet growth kinetics and pelletization behavior and the influence of operating variables by considering the complete size distributions is not convenient. Accordingly we make use of average pellet diameter (computed by using Equation 3-3) for further discussion.

## 4.2 Reproducibility of Pelletization Kinetics

A major obstacle that we have encountered during the initial stages of the project was reproducibility of batch pelletization results with Illinois #6. Figure 4.5 shows the batch pelletization results with Illinois #6 coal where average pellet diameter is plotted as a function

**(a) Coalescence**

**(b) Raspberry growth**

(a)        (b)
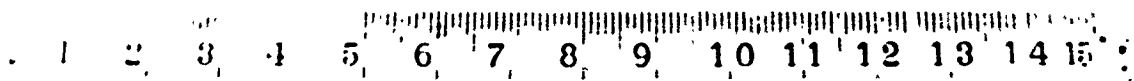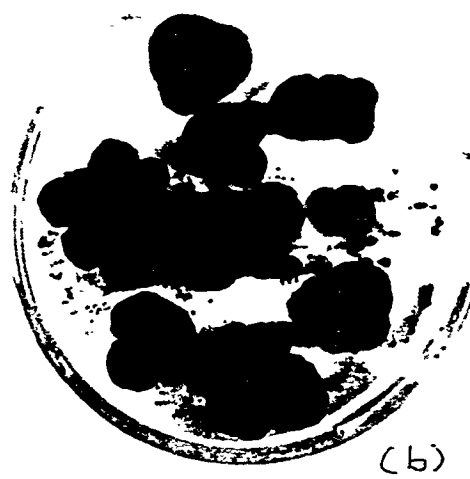
Figure 4.3. Schematic of formation and photographs of (a) spherical and (b) raspberry shaped pellets.
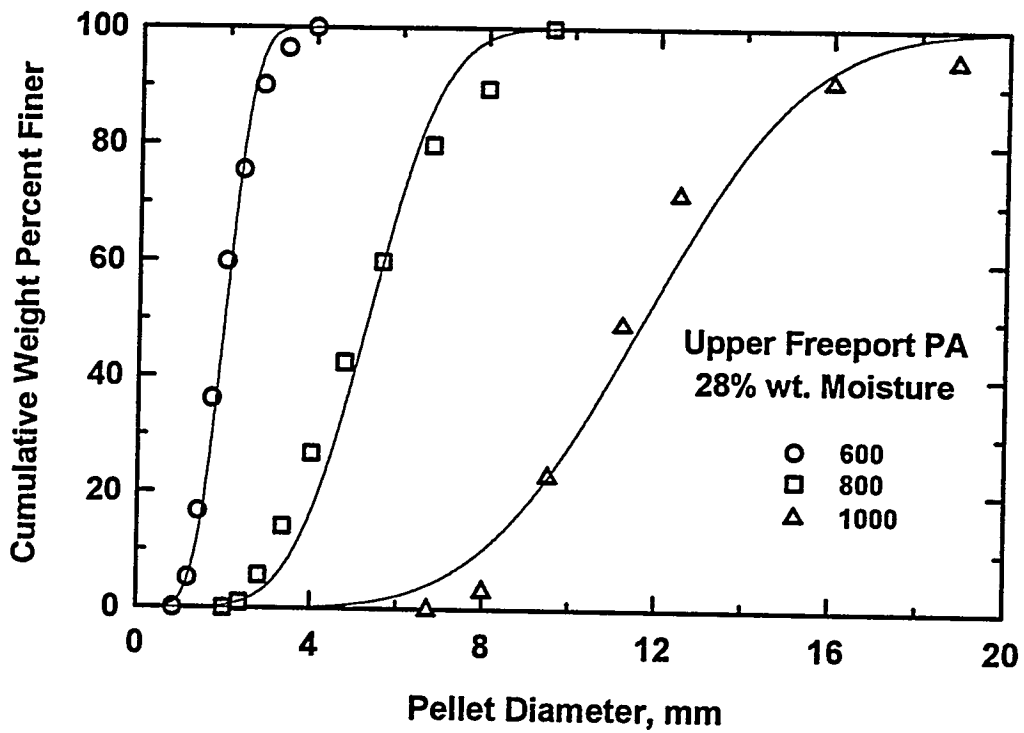
Figure 4.4. Size distribution of coal pellets at different stages of growth during a batch pelletization experiment with Upper Freeport PA fine coal sample.
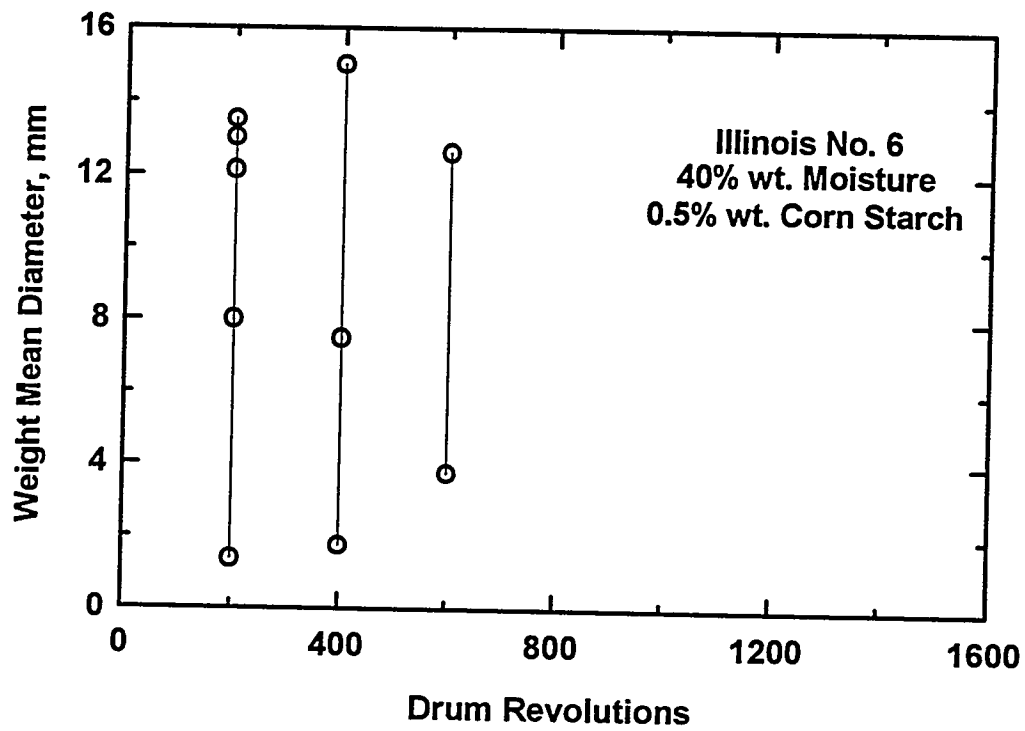
Figure 4.5. Reproducibility of coal pelletization experiments (Illinois #6 Sample).

of number of drum revolutions. This figure indicates the typical problem associated with the reproducibility of the experimental results. Using extreme precautions in conducting the experiments, we were able to improve the reproducibility of the results. However, we found that it is often necessary to repeat an experiment several times and take average of several measurements in order that any reasonable interpretation of the data becomes possible.experimental results. It took several months in resolving the reproducibility problem.

Because Illinois No. 6 coal could not be depended upon to maintain a consistent surface composition, despite precautions such as purging storage drums with argon, further reproducibility studies were conducted with Upper Freeport PA. Figure 4.6 demonstrates that reasonably good reproducibility was obtained at 26% and 30% added moisture. Each curve has a fairly narrow variation of the weight mean diameter. However, the 28% curve contains a critical region. Three distinct regions are observed on this curve.

1. The early part of the curve, where nucleation occurs predominantly, experiments show small variations in the weight mean diameter of the product pellets.

2. The transition region, where pellet growth by coalescence dominates, turns out to be the most critical region where there is a significant variation in the weight mean diameter and poor reproducibility.

3. Finally, in the ball growth region there tend to be small variations in the weight mean diameter.

The critical region, which exhibits large variability, is probably due to the extreme sensitivity of the pelletizing curve to any kind of experimental perturbation. For example, although the feed for pelletization is rubbed through a 6 mesh screen to fluff the moist feed, there must be some variation in the size of the minus 6 mesh product between different experiments. This

4-8
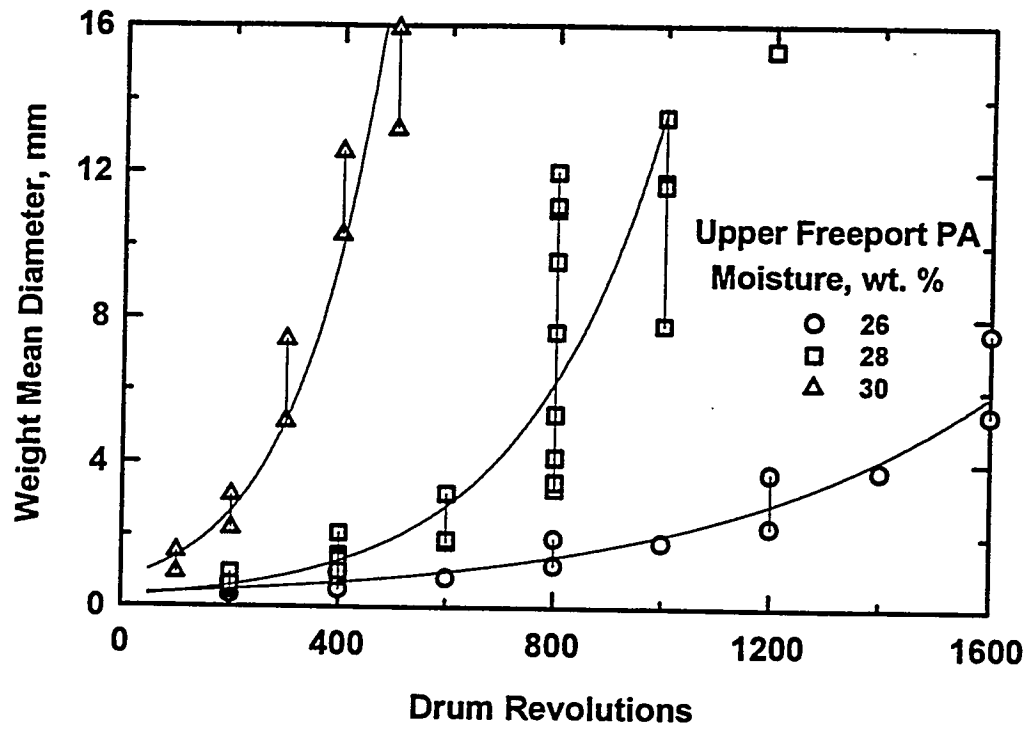
Figure 4.6. Reproducibility of coal pelletization experiments (Upper Freeport PA sample).

variation in feed moisture size could then alter the size distribution of the nuclei and resulting kinetics in the critical region.

All in all, we discovered that pelletization behavior of coal fines in general is highly sensitive to even minor variations in the coal characteristics especially changes (such as oxidation) taking place during storage and pellet feed sample preparation.

## 4.3 Influence of Feed Moisture

Figure 4.7 shows the effect of feed moisture on the pelletization behavior of Upper Freeport coal fines. As is normally observed, the rate of pellet growth increases with increased feed addition. As pointed out earlier, reproducibility was not very good and it often necessitated to conduct several experiments and take average values for the weight mean diameter before any meaningful trends could be established as was done in this case from the results in Figure 4.6. Of course it must be realized that the pelletization is possible only within the narrow range of moistures, i.e., between about 26 to 30 percent for Upper Freeport coal sample. Outside this range the feed gets either too dry to pelletize or too wet to make a green pellet.

Figure 4.8 shows the influence of the moisture content on the batch pellet growth behavior of Illinois No. 6 coal. Again, as expected the pellet growth rate increased with increased moisture addition. Nevertheless, the growth rate is found to be inductive and erratic, in that with low moisture addition, the pellets tend to grow slowly during the first several drum revolutions (induction) with clumping type growth at a later stage. We found that the overall growth behavior of Illinois #6 coal fines is fairly erratic in that the pellets obtained from this coal are not smooth like with others. This particular coal pelletized with some consistency in the range of 28 to 32 percent added moisture in March 1990. But, within two months, the same coal sample pelletized with moisture additions ranging from 34 to 38 percent added moisture.
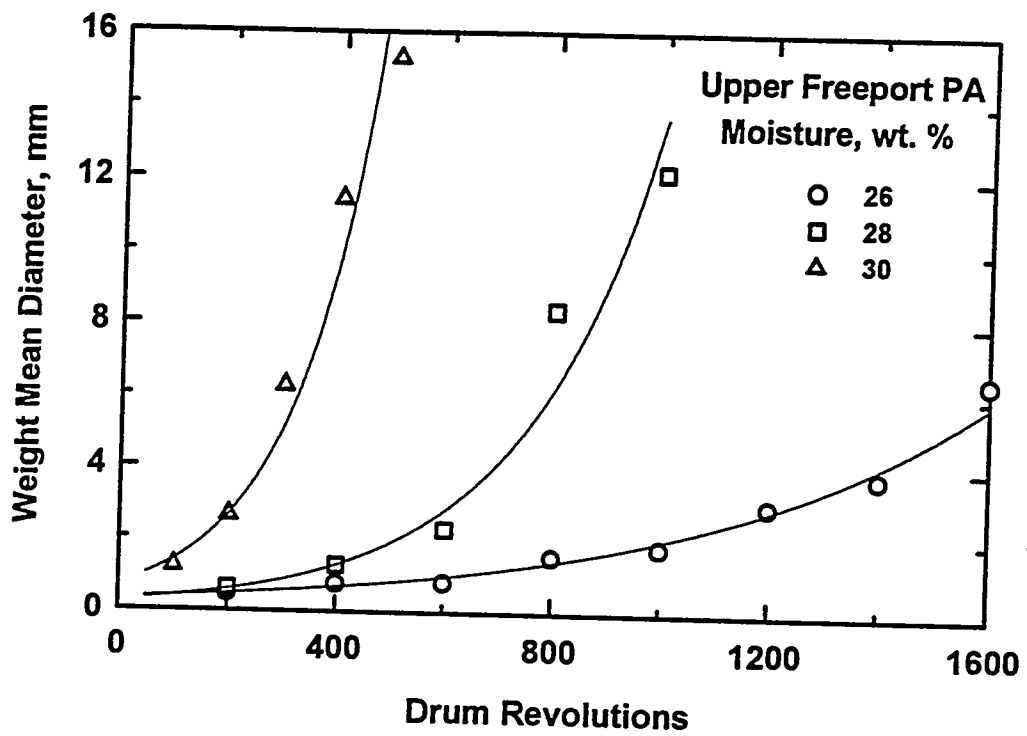
Figure 4.7. Effect of feed moisture content on the batch pelletization kinetics of the Upper Freeport PA sample.
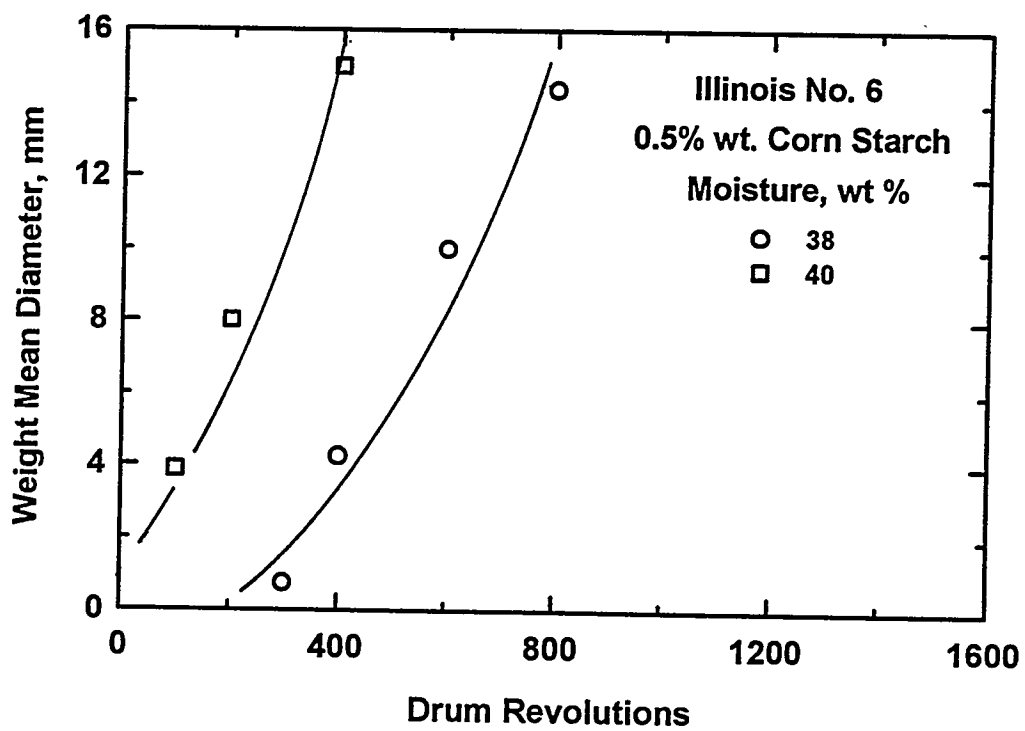
Figure 4.8. Effect of feed moisture content on the batch pelletization kinetics of the Illinois No. 6 sample.

By September 1990, the moisture requirements for pelletization had increased up to 42 to 44 percent added moisture (Figure 4.9).

Fuerstenau et al. [18] found that the surface pH of this coal sample had shifted significantly from a near-neutral (pH=6.5) to an acidic (pH=4) over a period of 4 months. They concluded that this pH shift was caused by auto-oxidation of ash-forming compounds including sulfur-compounds, despite the inert storage conditions. They also observed a shift in the hydrophobicity of this coal, towards increasing hydrophillicity. The pH shift is probably caused by auto-oxidation, caused by sulfur-bearing compounds reacting with atmospheric oxygen and residual moisture to form sulfuric acid. The oxygen could be retained within the pores of the coal and not displaced by argon when purging the stored samples. These kind of factors result in the erratic pelletization behavior by Illinois #6 sample.

In contrast to bituminous coals, the pelletization behavior of Bethlehem anthracite is more stable as evidenced by Figure 4.10. These curves exhibit small variability, throughout all different moisture additions. Again, increasing moisture addition increases the rate of growth. But the shape of the growth curves is substantially different from those for Upper Freeport and Illinois #6 coals. The only way we can explain this difference is in terms of the occurrence of different pellet growth mechanisms and this topic is addressed in the next section in more detail.

## 4.4 Influence of Addition of Dodecane

This section discusses the influence of addition of dodecane on the pellet growth behavior. Dodecane is used as a collector during flotation of fine coals. Kinetic data from the pelletization experiments on the four coal samples are presented in Figures 4.11 through 4.14. Results from Figures 4.11 and 4.12 for Upper Freeport and Pittsburgh #8 coals show that dodecane addition decreases their growth rate. Figure 4.13 for Illinois #6 does not show any definitive trend in
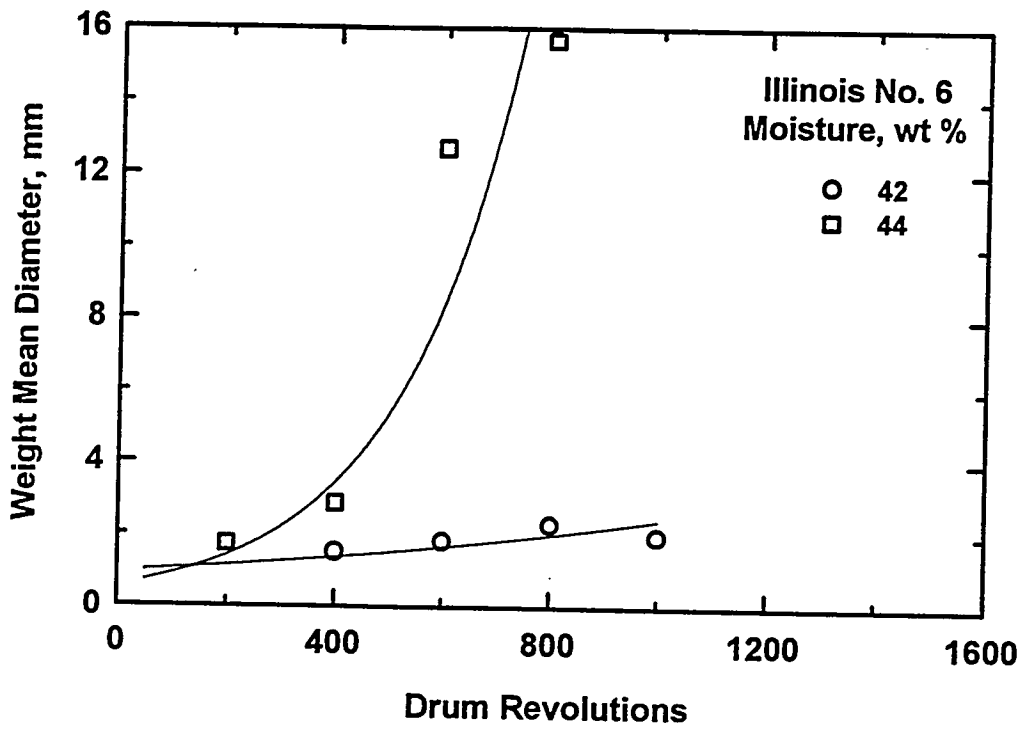
Figure 4.9. Moisture reproducibility problems with Illinois No. 6
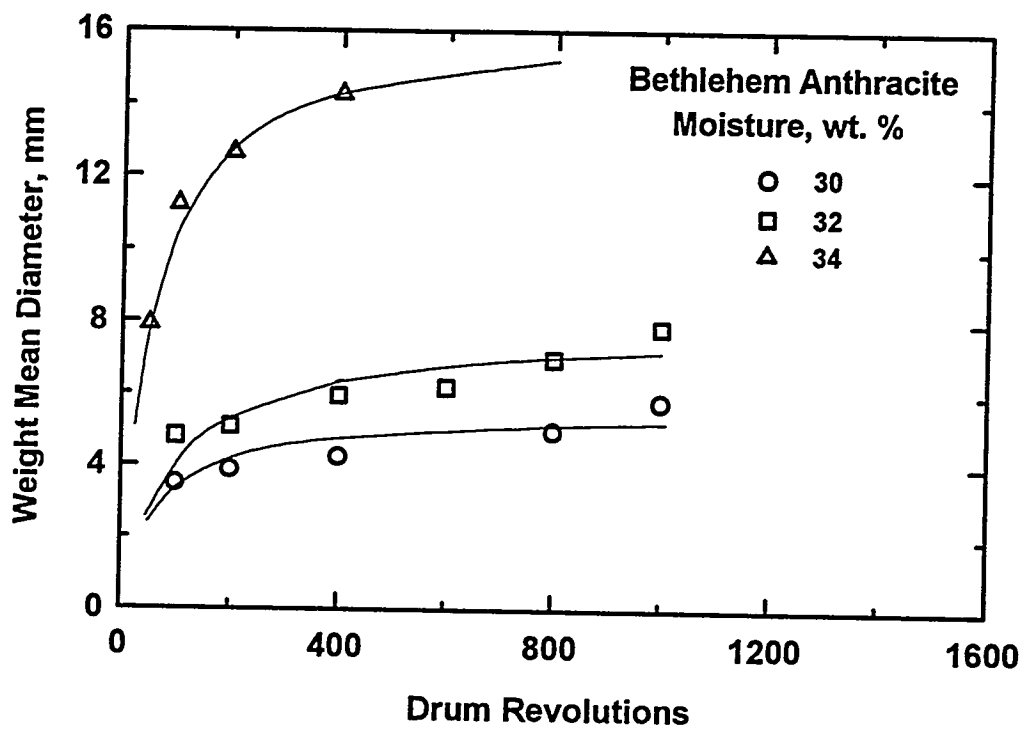
Figure 4.10. Effect of feed moisture content on the batch pelletization kinetics of the Bethlehem Anthracite sample.
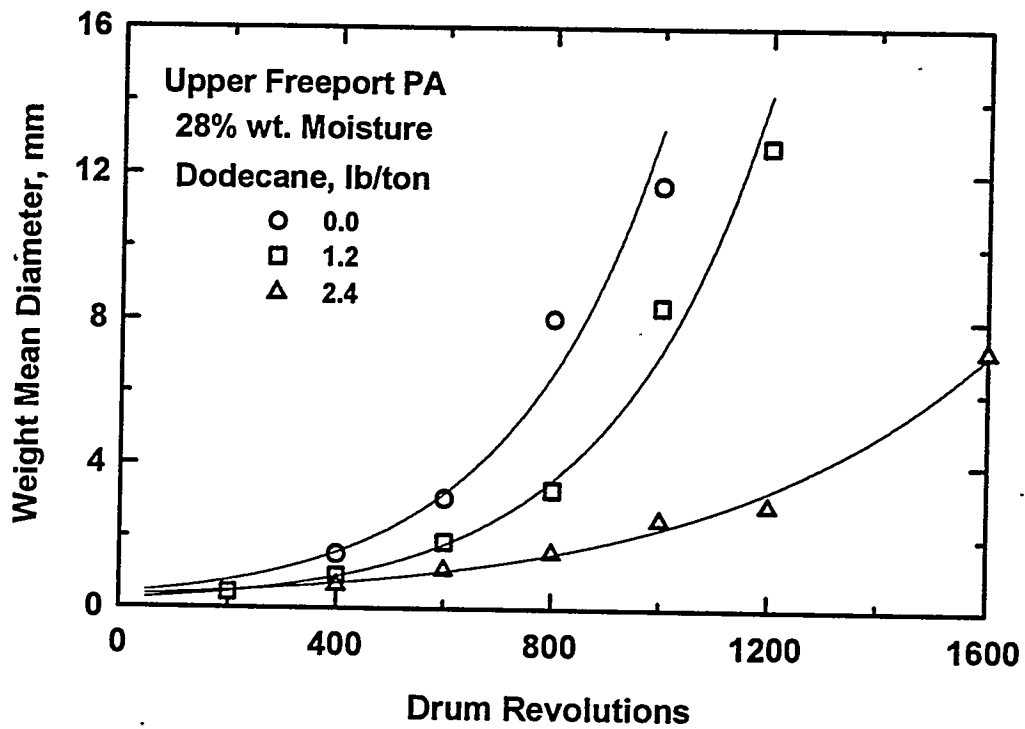
Figure 4.11. Influence of dodecane addition on the batch pelletization kinetics of the Upper Freeport PA sample.
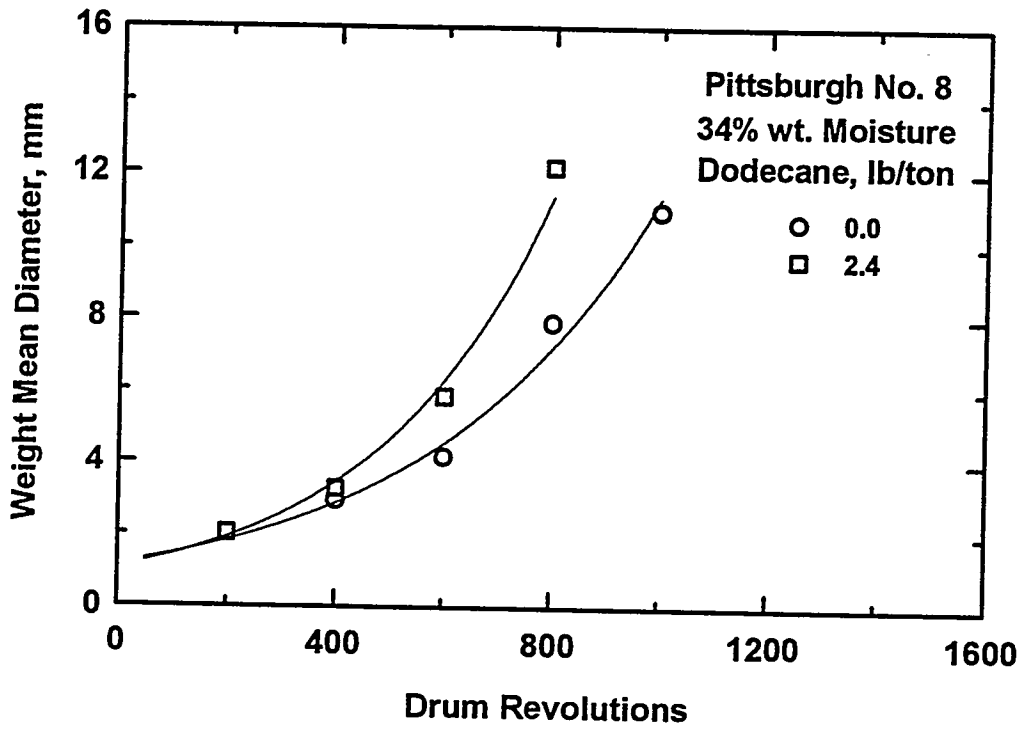
Figure 4.12. Influence of dodecane addition on the batch pelletization kinetics of the Pittsburgh No.8 sample.
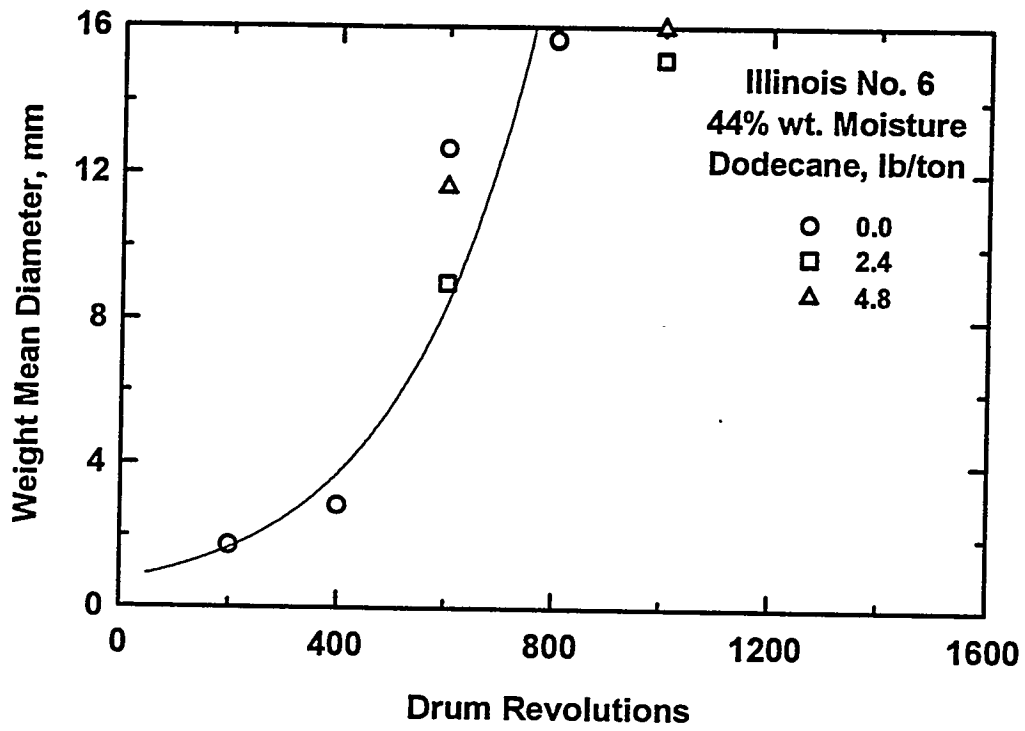
Figure 4.13. Influence of dodecane addition on the batch pelletization kinetics of the Illinois No.6 sample.
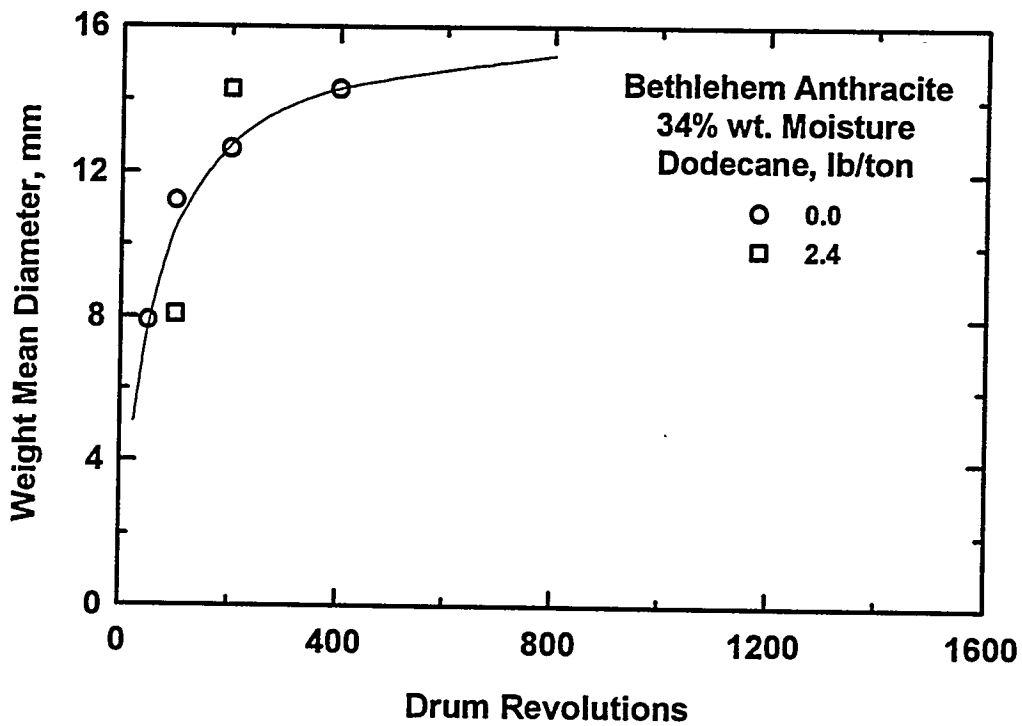
Figure 4.14. Influence of dodecane addition on the batch pelletization kinetics of the Bethlehem Anthracite sample.

the change in the pelletization behavior. Also, the pelletization behavior of Bethlehem anthracite is not influenced by the addition of dodecane.

We know that the addition of dodecane makes the surface of the coal matter more hydrophobic. Then we recognize that pendular and capillary forces are weaker in holding the particles within an agglomerate. Also when two agglomerates encounter each other, the efficiency of coalescence is also lowered. These two factors are expected to give rise to lower growth rates. While the pelletization behavior of Upper Freeport and Pittsburgh #8 coals (Figures 4.11 and 4.12) agree with the above explanation, Illinois #6 does not because of its erratic and unstable surface properties. In the case of Bethlehem anthracite, addition of dodecane does not change its already high hydrophobicity and as such does not lead to any change in the pelletization rates.

An analysis of the data and a formal interpretation of the role of dodecane on the pelletization behavior should be attempted especially in terms of the possible implications of ultimate analysis, functional groups, and structural effects of coals.

## 4.5 Effect of Addition of Dextrin

Influence of the addition of dextrin on the batch pelletization behavior of coal fines is presented in Figures 4.15 through 4.18. All the three bituminous coals (Figures 4.15, 4.16 and 4.17), indicate that the addition of can significantly accelerate the kinetics. For Upper Freeport PA coal, the effect of the acceleration is equivalent to up to 4% of added moisture (Figure 4.15) and is similar in magnitude for Pittsburgh No. 8 (Figure 4.16) and Illinois No. 6 (Figure 4.17). It is known that dextrin causes the surface of coal to become hydrophilic. This increased hydrophillicity is expected to give rise to higher efficiency of coalescence and more stable and stronger agglomerates both factors leading to increased growth rate. Our experiments clearly
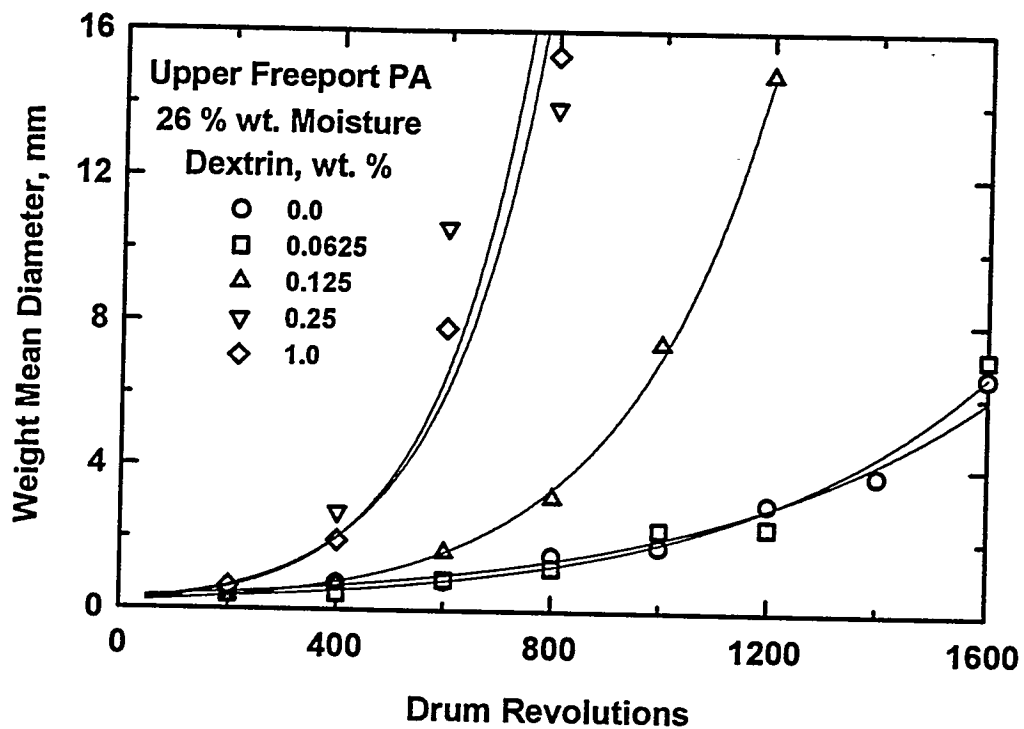
Figure 4.15. Effect of dextrin addition on the batch pelletization kinetics of the Upper Freeport PA sample.
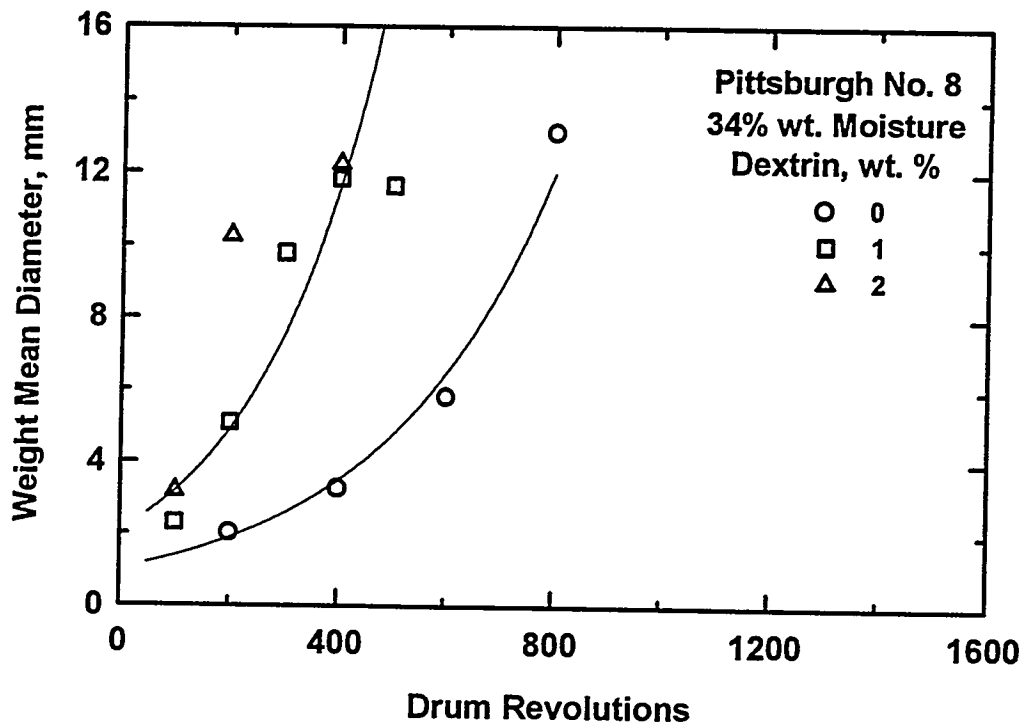
Figure 4.16. Effect of dextrin addition on the batch pelletization kinetics of the Pittsburgh No.8 sample.
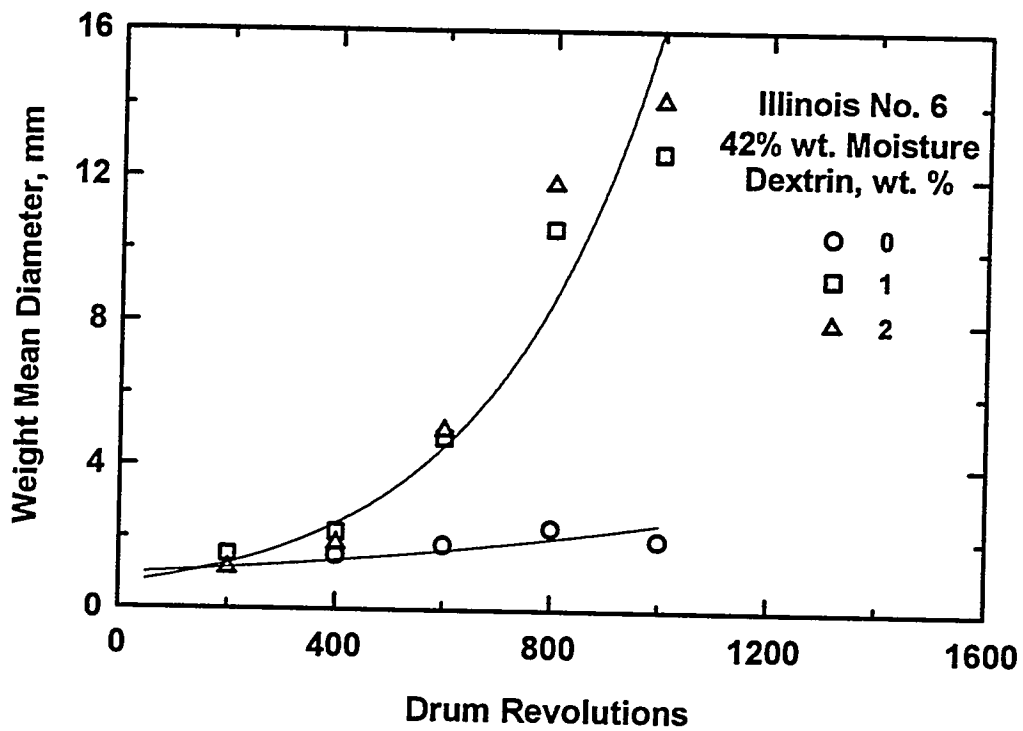
Figure 4.17. Effect of dextrin addition on the batch pelletization kinetics of the Illinois No.6 sample.
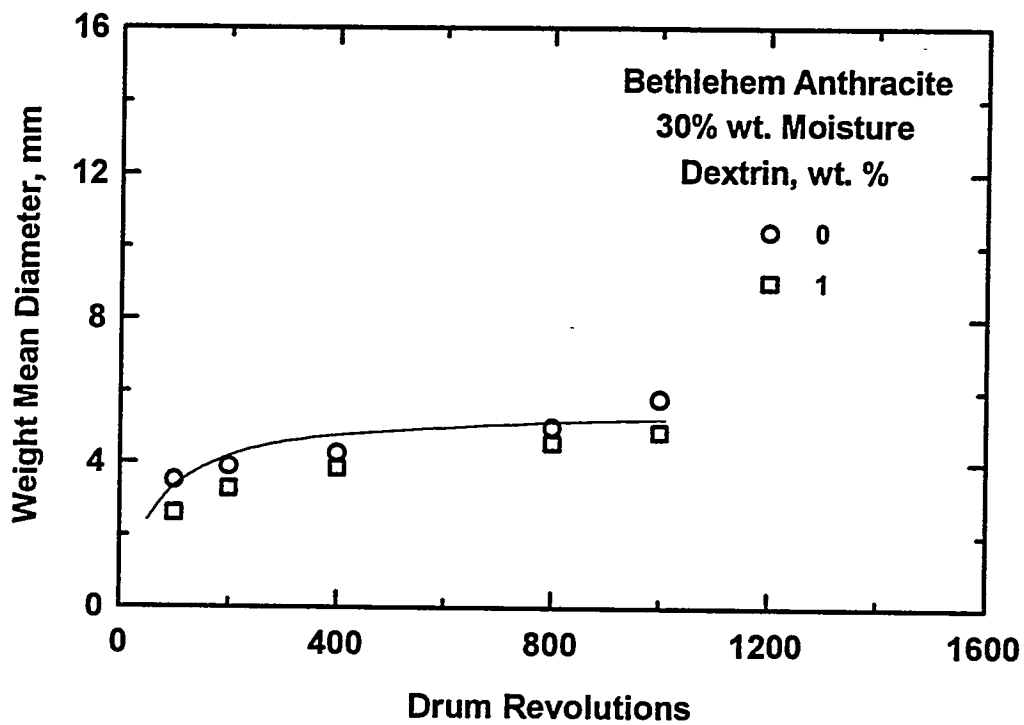
Figure 4.18. Effect of dextrin addition on the batch pelletization kinetics of the Bethlehem Anthracite sample.

indicate that the increase in the growth rate reaches an asymptotic limit at about 1% addition of dextrin.

Bethlehem anthracite does not show any change in its pellet growth behavior due to the addition of dextrin (Figure 4.18). Since this sample of anthracite is relatively pure, with only a very small percent of hydrogen and volatile compounds (Table 3.2), it is expected that there would be few surface sites for the adsorption of dextrin. Conversely, since the three bituminous coal samples do contain significant amounts of volatiles and hydrogen, there would be many more surface sites for dextrin adsorption and increased hydrophillicity.

### 4.5.1 Film Flotation Results

This section discusses our efforts to characterize the causes for the effect of dextrin by making use of a relatively new technique called film flotation developed by Hornsby and Leja (1984) and Fuerstenau et.al. (1990). The film flotation experiments were conducted on the as received and dextrin treated samples of Bethlehem anthracite, Illinois No.6. Pittsburgh No. 8, and Upper Freeport, PA coals and the results are presented in Figures 4.19 through 4.22.

The film flotation technique consist of placing a monolayer of particles onto the surface of a liquid of a known surface tension. Depending on the surface energy of the particles, the particles separate into a sink or float fraction. As the surface tension is varied, the particles are partitioned into lyophobic (float) or lyophillic (sink) fractions. The weight percent of the lyophobic fraction is then plotted as a function of the solution surface tension to produce a "wetting tension distribution diagram" and the mean surface tension and contact angle are calculated by following the procedure recommended by Fuerstenau et.al. The mean surface tension and the contact angle are good measures of wettability and as the particle wettability is increased the mean surface tension increases and the contact angle decreases.

4-25
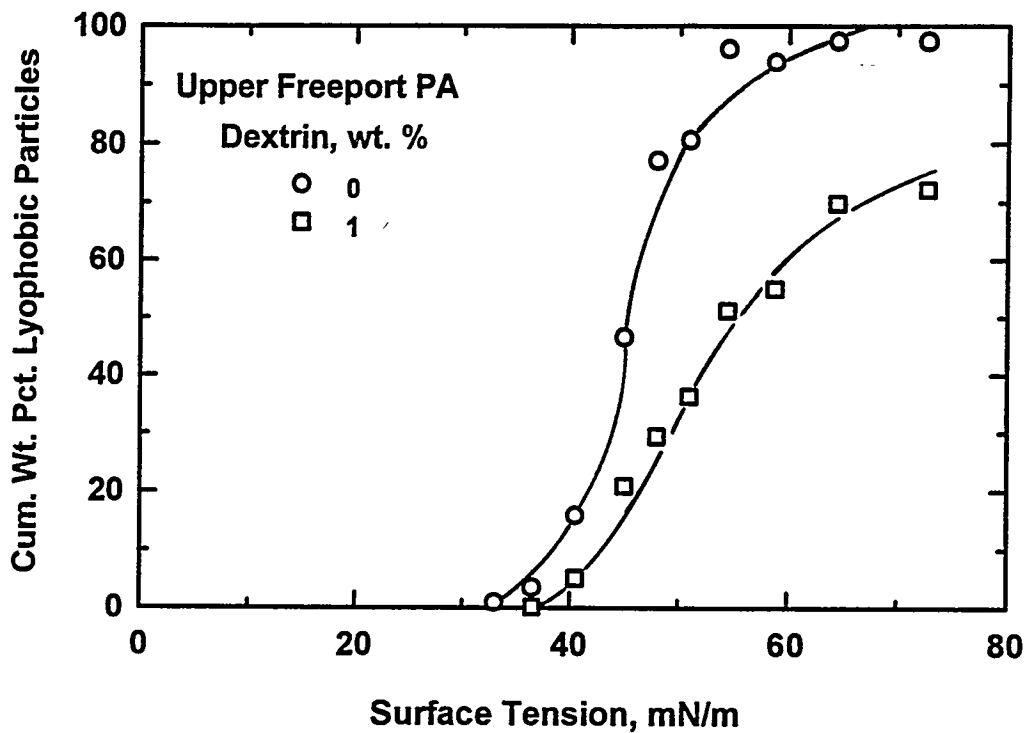
Figure 4.19. Cumulative weight fraction of lyophobic particles as a function of their wetting surface tension for Upper Freeport PA coal fine sample.
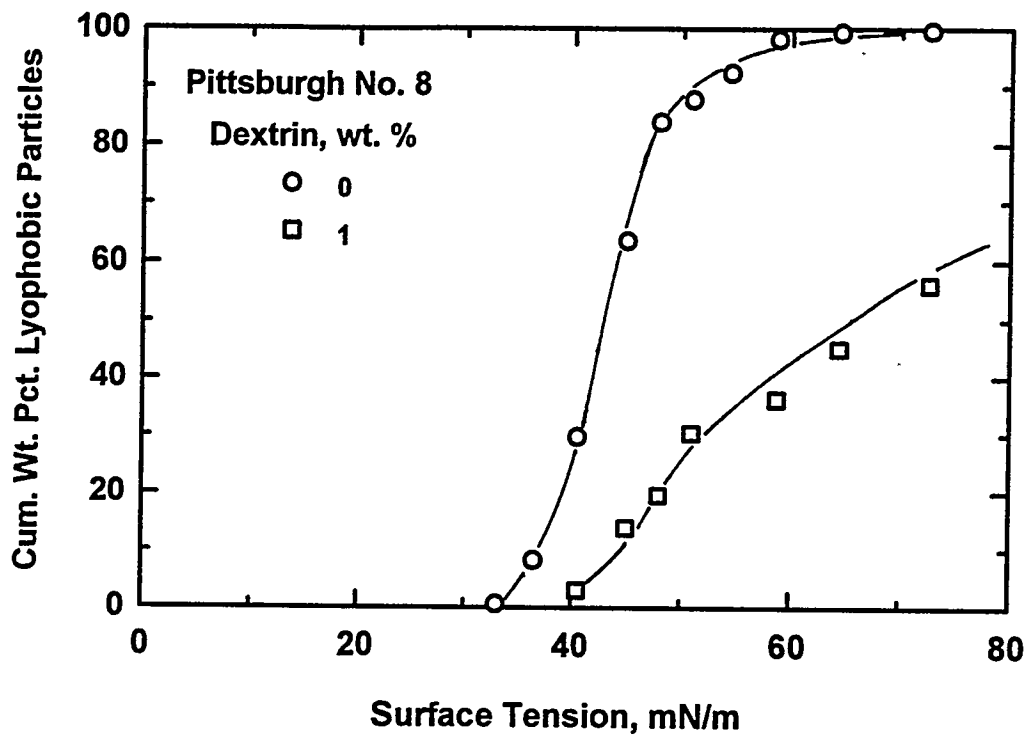
Figure 4.20. Cumulative weight fraction of lyophobic particles as a function of their wetting surface tension for Pittsburgh No. 8 coal fine sample.
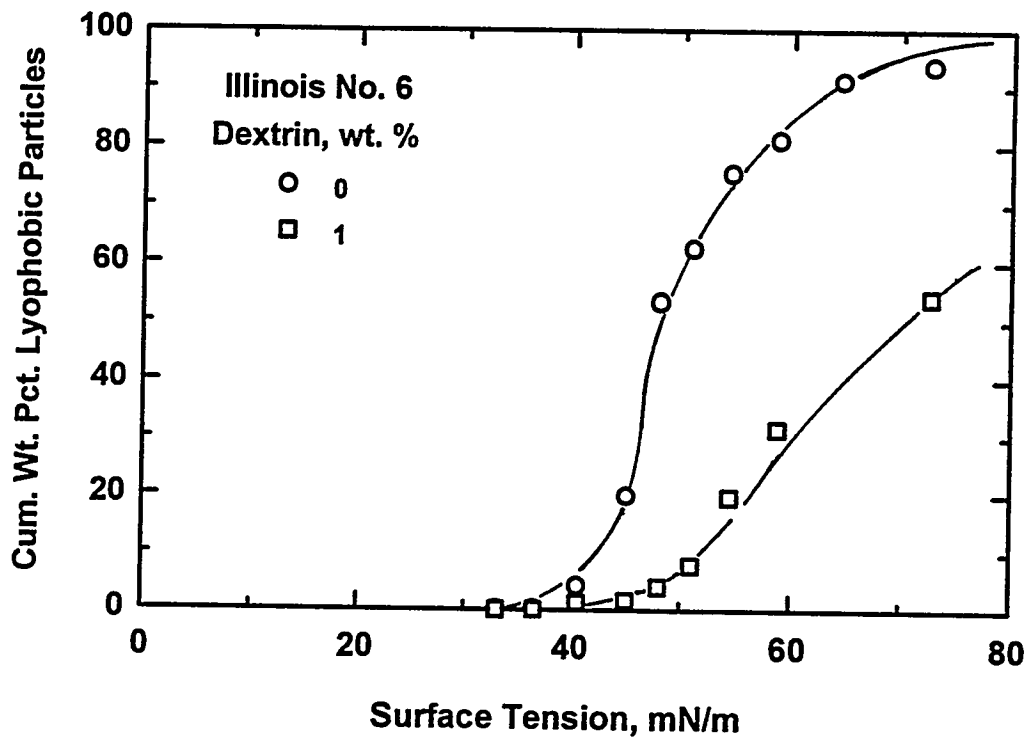
Figure 4.21. Cumulative weight fraction of lyophobic particles as a function of their wetting surface tension for Illinois No.6 coal fine sample.
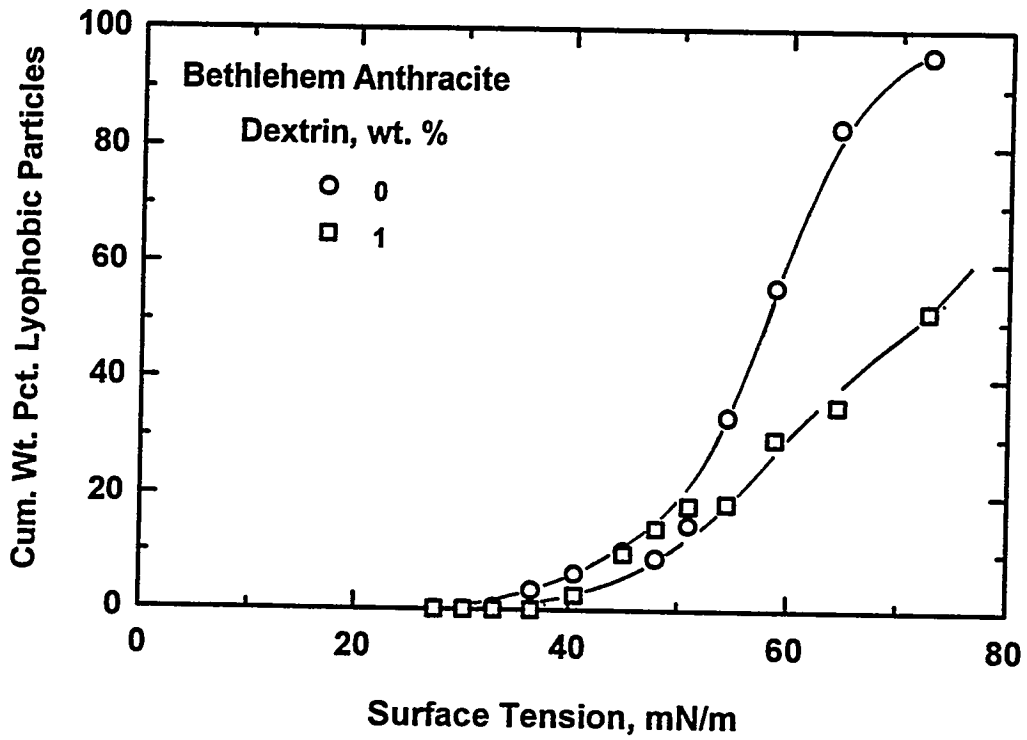
Figure 4.22. Cumulative weight fraction of lyophobic particles as a function of their wetting surface tension for Bethlehem anthracite coal fine sample.

The present film flotation experiments were carried out by sprinkling a monolayer of 100X150 mesh particles on the surface of an aqueous methanol solution and then separating the sink and float fractions. The aqueous methanol solutions ranged from pure water (surface tension 72.8 mN/m) to pure methanol (surface tension 22.4 mN/m). This set of experiments was conducted at $20^0C$. The surface tension of the aqueous methanol solutions was determined by using a Fisher Tensiometer. The cumulative lyophobic distributions for these four coals are presented in Figures 4.19 through 4.22. The wetting parameters, (i.e., mean surface tension and contact angle) derived from the film flotation results are given in Table 4.1. These results show that the cumulative distribution curve of the dextrin-treated coal shifts to a higher wetting surface tension as compared with that of the as-received samples. All these data clearly indicate that the percentage of hydrophobic surface sites decrease with the addition of dextrin. Also, as expected, the addition of dextrin increases the mean surface tension values and decreases the contact angle, a clear indication of increased wetting (or increased hydrophillicity).

There are some remarkable observations in these pelletization kinetic data. First, the pelletization rate of Bethlehem anthracite does not change with the addition of dextrin in spite of the fact that there is a substantial increase in the surface wettability. In contrast all the other three coals exhibit noticeable increase in the growth rates with the addition of dextrin which caused increased wettability. We feel that the structure of coal and such functional groups as carboxylic and phenolic groups play a major role in actually determining the pelletization behavior in conjunction with the surface wettability in itself. An interpretation of these factors should be attempted in the future work.

Table 4.1. Contact angles for as-received and dextrin treated coal fines.

| Coal | Modification | mN/m | $\Theta$, deg |
|---|---|---|---|
| Bethlehem | As-Received | 57.5 | 43 |
| Anthracite | 1% Dextrin | 72.0 | 10 |
| | | | |
| Illinois | As-Received | 49.0 | 57 |
| No. 6 | 1% Dextrin | 70.5 | 14 |
| | | | |
| Pittsburgh | As-Received | 42.5 | 69 |
| No. 8 | 1% Dextrin | 68.0 | 22 |
| | | | |
| Upper | As-Received | 45.0 | 64 |
| Freeport, PA | 1% Dextrin | 55.0 | 47 |

## 4.6 Influence of Coal Weathering

Figure 4.7 showed the pelletization behavior of as-received Upper Freeport coal fines with different addition of moisture contents. Like most coals, this coal could be pelletized only over a narrow range of moisture additions of 25 to 32 percent by weight (dry basis). With lower moisture addition, the Upper Freeport coal exhibits a much longer induction period followed by a rapid, clumping type growth.

Next set of experiments were carried out on Upper Freeport coal sample that was stored for 13 weeks in open drums and subjected to weathering. Pelletization of this weathered coal with 32 percent moisture was found to be extremely slow. Figure 4.23 presents the growth curves for the as-received and weathered samples of Upper Freeport coals. The behavior of weathered coal is similar to that of the as-received coal with low moisture addition.

Subsequently, we conducted tests on the weathered sample by increasing the moisture addition to a substantial amount, i.e. up to 36 percent. These results are also shown in Figure 4.23 in which the growth curve for the as-received sample is included for comparison purposes. It is clear that the growth rate could be increased by increasing the moisture addition. We believe that the differences in the pelletization behavior of the as-received and weathered coals must result from surface oxidation and the resulting changes in wetting behavior.

In order to see if the weathering influenced the growth mechanisms during the pelletization process, we have plotted the size distributions of pellets (with 10 mm weight mean diameter) produced with the as-received and weathered samples. These results are given in Figure 4.24. The pellet size distribution is not changed in spite of the inductive type of growth with the weathered sample. This indicates that the mechanisms of pellet growth are not altered by weathering.
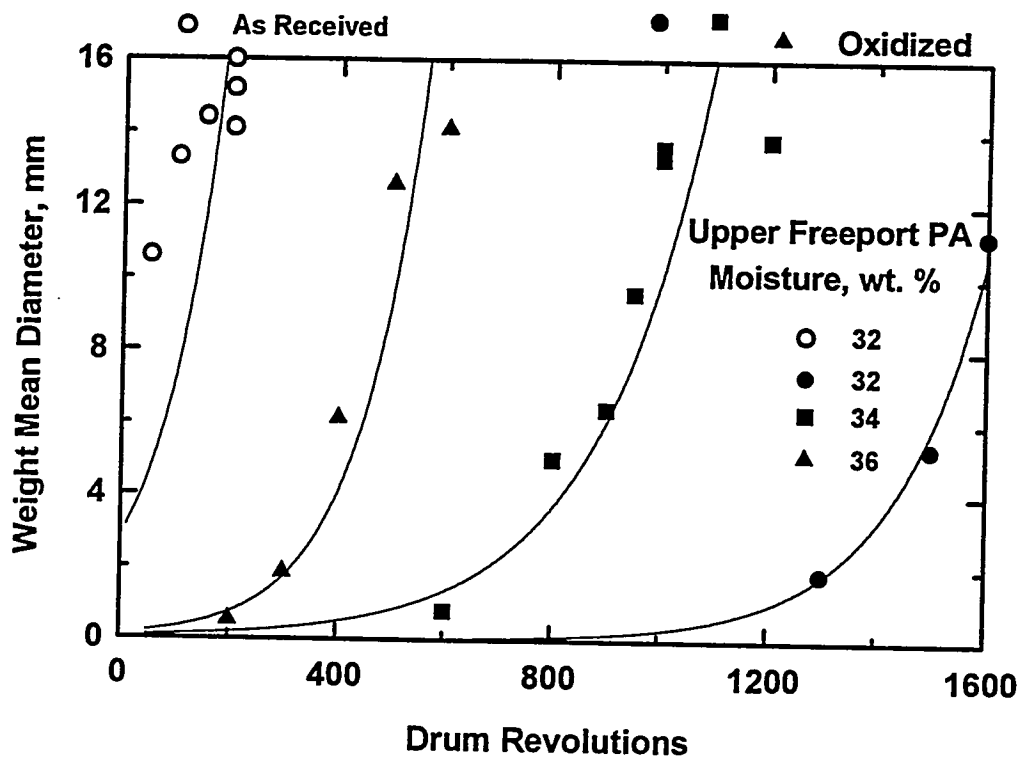
Figure 4.23. Influence of weathering oxidation of on the pelletization behavior of Upper Freeport PA coal fine sample.
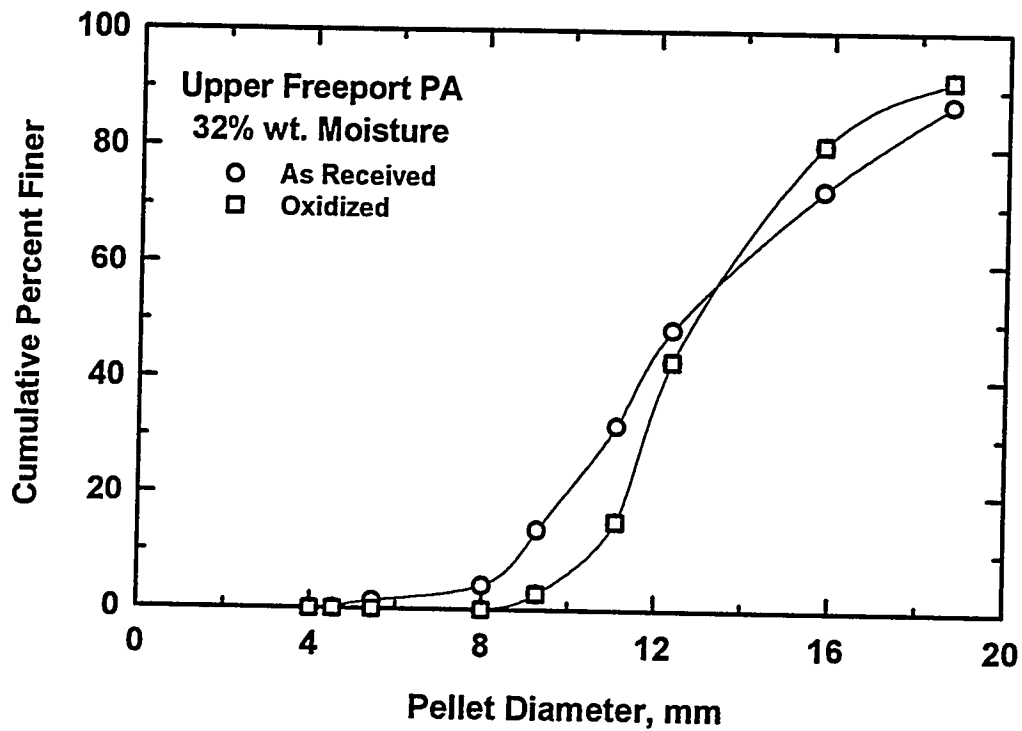
Figure 4.24. Size distribution of agglomerates produced from as-received and oxidized samples of Upper Freeport PA coal fines.

It is well known that during storage, coal samples undergo weathering essentially causing surface oxidation. We believe that the differences in the pelletization behavior of the as-received and weathered coals must result from this surface oxidation. Surface oxidation increases wettability of the coal which in turn can increase the moisture requirement for pelletization. It is also believed that oxidized coal immobilizes substantial quantities of moisture thus less moisture becomes available for pelletization. One method to check this hypothesis is valid is to determined the air dried moisture of as received and weathered coals and then determine the free moisture of pellet feeds. This free moisture shall correspond to the moisture available for pelletization.

## 4.7 Control of Coal Pellet Growth Process

Surging is a common phenomenon in conventional pelletization circuits (see also Section 6). This is reported to be caused by uncontrolled occurrence of various mechanisms of pellet formation and growth. It is desirable to control the pelletization process by deliberately changing the extent of contribution from each mechanisms through redesign of the circuit or proper control of the operating conditions. In this context, we have found that nuclei growth by rapid coalescence, as opposed to coalescence followed by compaction, results in raspberry type growth (Figure 4.3). The raspberry shape is not as desirable as spherical pellets.

We have conducted several qualitative experiments to analyze the raspberry type growth and see if it could be prevented. Normally coalescence involves two pellets at a time. But in the case of raspberry type growth a number of pellets stick together at a time resulting in a growth as represented in Figure 4.3. The cause for this kind of growth, we believe, is related to the excess stickiness on the pellet surface, lower deformability, and above all the light weight of the

coal agglomerates. Accordingly, we decided to try two methods which are expected to reduce the rapid coalescence.

The first method was aimed at reducing the stickiness of the pellet surface. As the particle size becomes smaller the pellets formed out of that becomes smoother. This results in a larger contact during collisions and in turn increases the possibility of coalescence. Hence, we tried to study the effect of feed size distribution on the pelletization behavior of coal and found that coarser distribution would reduce coalescence and result in spherical pellets. It was found that coarser distributions would reduce the raspberry type of growth. The second method was based on the reduction in the coalescence mechanism by deliberately promoting layering mechanism. We also found that coarse powders tend to layer more readily than finer coals. The fine coals tend to nucleate and give rise to raspberry growth.

Our experiments indicate that the feed size distribution is critical in determining the growth mechanism and the shape of the pellets. It may not always be possible to know the size distribution of fine coal feed for pelletization, because it may come from any number of sources such as (i) the mine site, (ii) the refuse pond, or (iii) the preparation plant. For example, the coal fines recovered from the refuse ponds may contain as much as 80 percent -325 mesh particles whereas the coal sample from a preparation plant may be somewhat coarser. If the coal is already in the finer than desired form there is no way of getting a coarse feed. Under such conditions layering will be the most appropriate method for prevention of raspberry type growth. In general, it is recommended to generate as coarse a size distribution as possible by blending fine and coarse coals.

# 5.0 MATHEMATICAL MODELING OF COAL PELLETIZATION PROCESS

In Chapter 4 we have discussed the influence of such operating variables as feed moisture content and additive content on the batch pelletization behavior of coal fines in a qualitative manner. In this chapter we shall attempt to develop suitable mathematical models for describing the coal pelletization kinetics.

## 5.1 Mathematical Modeling of Pelletization Process

The process of pelletization consists of tumbling moist fines in a rotating drum. Pellet size distributions are determined at various stages of the pellet growth and the average pellet diameter is then presented as a function of number of drum revolutions. Such a curve is called pellet growth kinetic curve. Figure 5.1 shows a typical growth curve form an experiment involving pelletization of taconite concentrates [54, 55]. This curve shows a characteristic sigmoidal shape. Kapur and Fuerstenau [31] also observed such a shape with the pelletization of pulverized limestone and suggested that the pelletization process could be recognized to proceed through three stages of growth: (i) nuclei, (ii) transition, and (iii) ball growth region.

Earlier, careful tracer studies were conducted to identify the mechanisms of agglomerate formation and growth and it was discovered [55] that the major mechanisms are (i) coalescence, (ii) breakage, and (iii) layering which have been depicted in Figure 2.2. Based on physical reasoning and extensive experimental observations, it is now agreed that the agglomerate growth during the nuclei-growth region occurs primarily by coalescence and during the ball growth region by breakage and layering. During the initial nuclei-growth stage, highly porous nuclei are generated by pendular bonds between loosely held moist particles due to the presence of water between the irregularly packed network of particles. On tumbling this moist charge in the
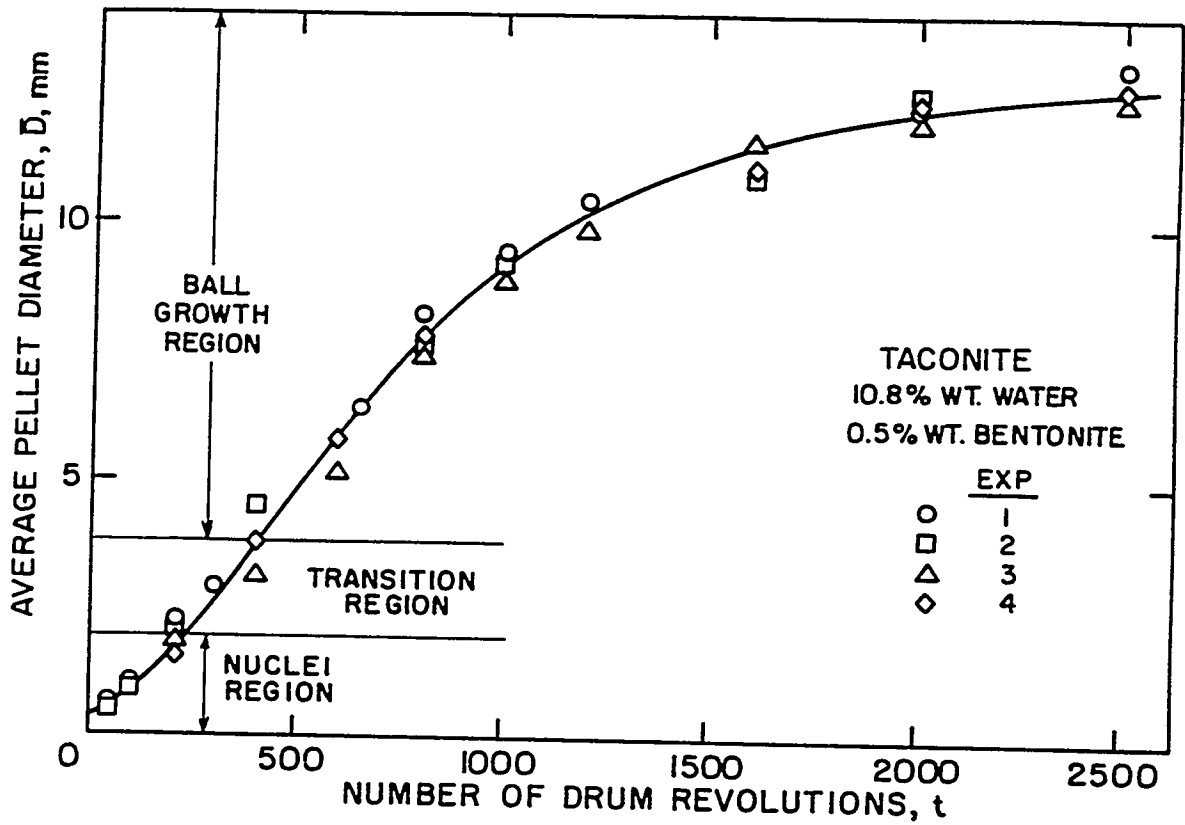
Figure 5.1. Typical pelletization curve for a taconite iron ore concentrate [53].

drum, the particles within the nuclei rearrange and pack together and form into stable porous spherical nuclei [31, 54, 55] because of capillary attraction. On further tumbling of the moist charge, the nuclei collide with one another, with some of the collisions tending to form a capillary neck and resulting in successful coalescence.

Mathematical models based on population balance models (PBM) have been developed earlier to describe the pellet growth process by one or more of mechanisms of agglomeration [32, 51, 55]. Equation 5-1 constitutes a generalized PBM equation.

$$\frac{\partial}{\partial t}[n(\delta,t)] + \frac{\partial}{\partial \delta}[k_L\, n(\delta,t)] + \bar{D} - \bar{A} = 0 \qquad [5\text{-}1]$$

where $n(\delta,t)d\delta$ is the number of agglomerates with pellet diameter in the interval $(\delta,\delta+d\delta)$ at time, $t$ and the symbols $k_L$, $\bar{D}$ and $\bar{A}$ describe respectively the layering rate parameter and the rate of appearance and disappearance of agglomerates due to the occurrence of other agglomeration mechanisms. Specific expressions for these appearance and disappearance terms need to be derived based on the mechanisms that are considered to be prevalent.

## 5.2. A Mathematical Model for Nuclei-Growth Region Pelletization

As pointed out above, the pellet growth mechanism during nuclei-growth region is coalescence. But, as will be shown in the final report, the weight-mean diameter, $\bar{D}_w$, of agglomerates growing by random coalescence follows an exponential growth pattern given by the following equation:

$$\frac{\partial}{\partial t}[n(\delta,t)] + \frac{1}{N(t)}\int_0^\infty k_C\, n(\delta,t)\, n(\alpha,t)\, d\alpha - \frac{1}{2N(t)}\int_0^\beta k_C\, n(\alpha,t)\, n(\beta,t)\, d\alpha = 0 \qquad [5\text{-}2]$$

where $N(t) = \int_0^\infty n(\delta,t)\,d\delta$ and $\delta^3 = \alpha^3 + \beta^3$; and $k_c$ is the coalescence rate parameter. When the coalescence rate parameter is not dependent on the size of agglomerates that are undergoing coalescence, then the process is called random coalescence and the Equation 5-2 can be readily solved [31, 32, 51] obtaining Equations 5-3 and 5-4 for the weight-size distribution and weight-mean diameter of the agglomerates:

$$W(\delta) = 1 - \exp[-(\delta/\bar{D}_w)^3] \qquad\qquad (5\text{-}3)$$

and

$$\bar{D}_w = \bar{D}_w(0)\,\exp(k_c t) \qquad\qquad (5\text{-}4)$$

where $W$ is the cumulative mass fraction of pellets smaller than the pellet diameter $\delta$ (in mm). It has been shown that size distribution given by Eq. 5.3 is normalizable as shown in Eq. 5.5 in the following. In other words the cumulative size distribution becomes pseudo-time independent. Such distributions are called self-preserving [31, 32, 51] which can be expressed as

$$W(\eta) = 1 - \exp[-\eta^3] \qquad\qquad (5\text{-}5)$$

where $\eta = \delta/\bar{D}_w$.

Now we see that Equation 5-4 demands that the weight mean diameter changes exponentially with time. An assessment of the results shown in Figures 5-2, 5-3 and 5-4, shows that the Upper Freeport and Pittsburgh #8 coals follow this prediction fairly closely, while exhibit nuclei type growth is not observed with the Bethlehem coal. Indeed our experimental observations substantiate this observation in that the more hydrophobic anthracite particles do not go grow by random coalescence.
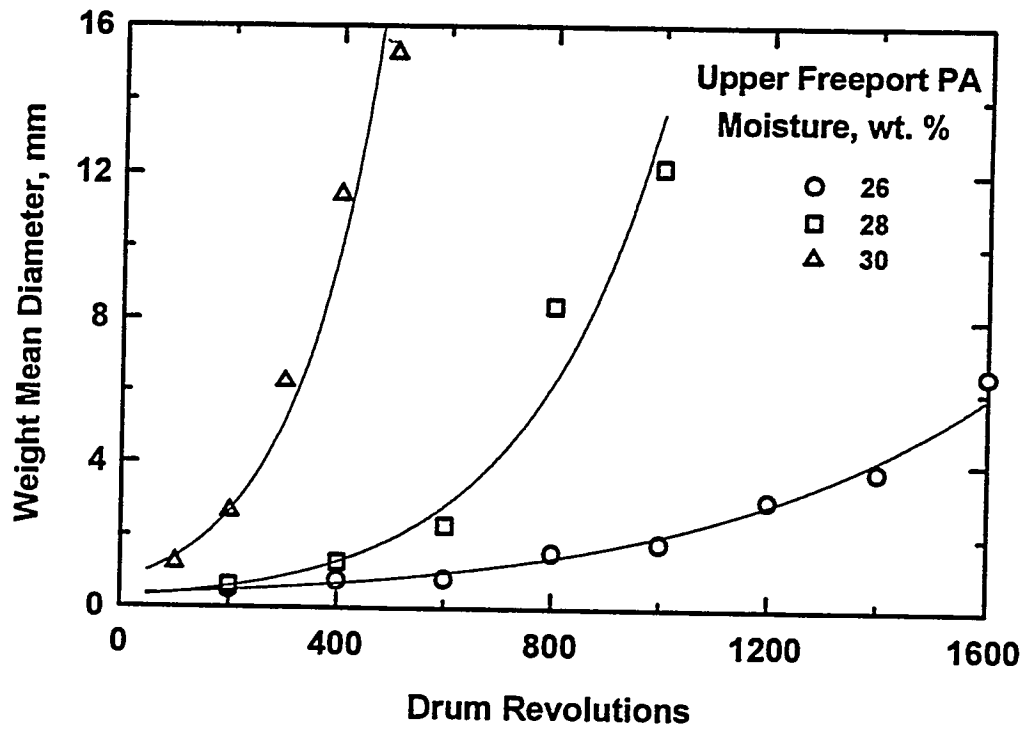
Figure 5.2. Effect of feed moisture content on the batch pelletization kinetics of the Upper Freeport PA sample (same as Figure 4.6). The solid lines in the figure show the exponential type of growth predicted by random coalescence model of Eq. 5.2.
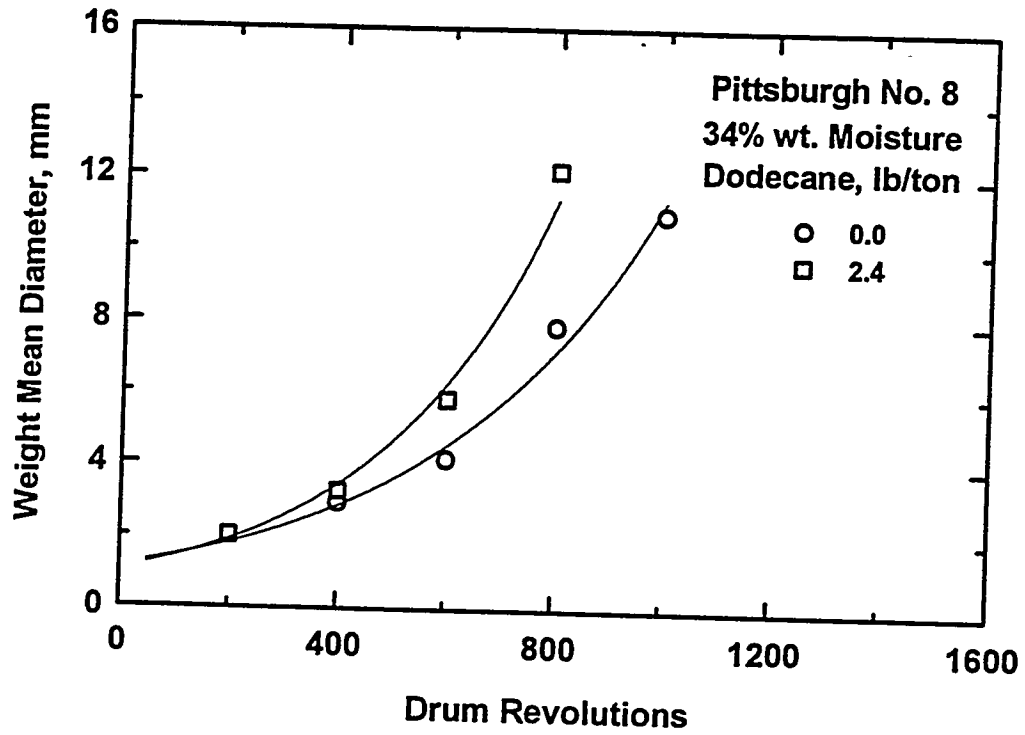
Figure 5.3. Effect of dextrin addition on the batch pelletization kinetics (same as Figure 4.12).
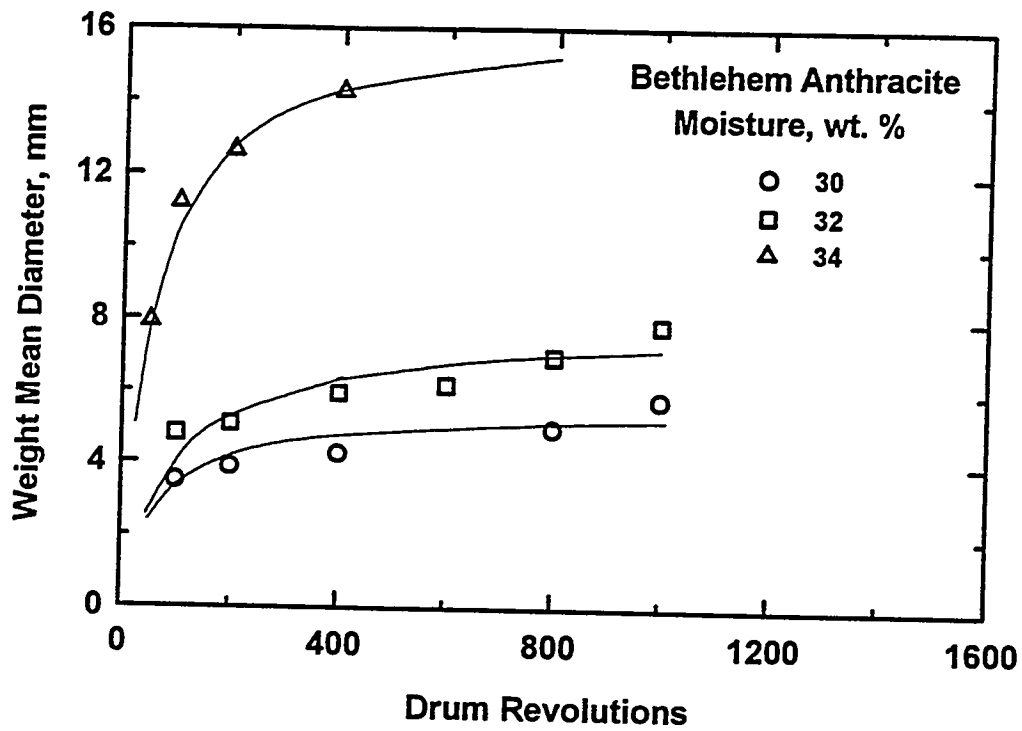
Figure 5-4. Effect of feed moisture content on the batch pelletization kinetics of the Bethlehem Anthracite sample (same as Figure 4-10).

In order to test the above theoretical predictions quantitatively, we tried to curve fit the experimental data from Figures 5.2, 5.3 and 5.4 to Eq. 1 and found better than 98 percent agreement with the theory for the Upper Freeport and Pittsburgh coals. The results are shown in Figures 5.2 and 5.3 where the solid curves correspond to Eq. 5-4. As can be seen from Figures 5.5 to 5.8 both the cumulative and the normalized distributions are in perfect agreement with the theoretical predictions (solid lines in the figures) as required by Eqs. 5-4 and 5-5. We also found that the agreement between the theory and experiment for the size distributions is better than 95 percent. While the Upper Freeport and Pittsburgh coals showed agreement, Bethlehem coal does not follow the random coalescence model as evidenced by Figure 5.4 for the growth curve and Figure 5.9 for the pellet size distribution.

## 5.3 Breakage and Layering Process

It is expected that the breakage and layering mechanism is predominant during the ball-growth region of a batch agglomeration process. We shall develop a mathematical model and compare the theoretical predictions with the experimental results.

Figure 5.10 shows specific interpretation of the breakage and layering process. In other words, we assume that the agglomerates of a given size can either break completely into extreme fragments or undergo a certain amount of abrasion. The resulting fines are expected to be layered instantaneously onto the surviving pellets. Further, we propose that the rate of breakage of agglomerates into small fines is inversely proportional to its diameter. In other words, the smaller pellets are more likely to be crushed into extreme fines at a higher rate than the larger ones. As per the layering, we make the standard assumption that the rate of increase of the mass of an agglomerate is proportional to its exposed surface area which in turn signifies that the rate of change of diameter of an agglomerate due to layering is constant. Making use of
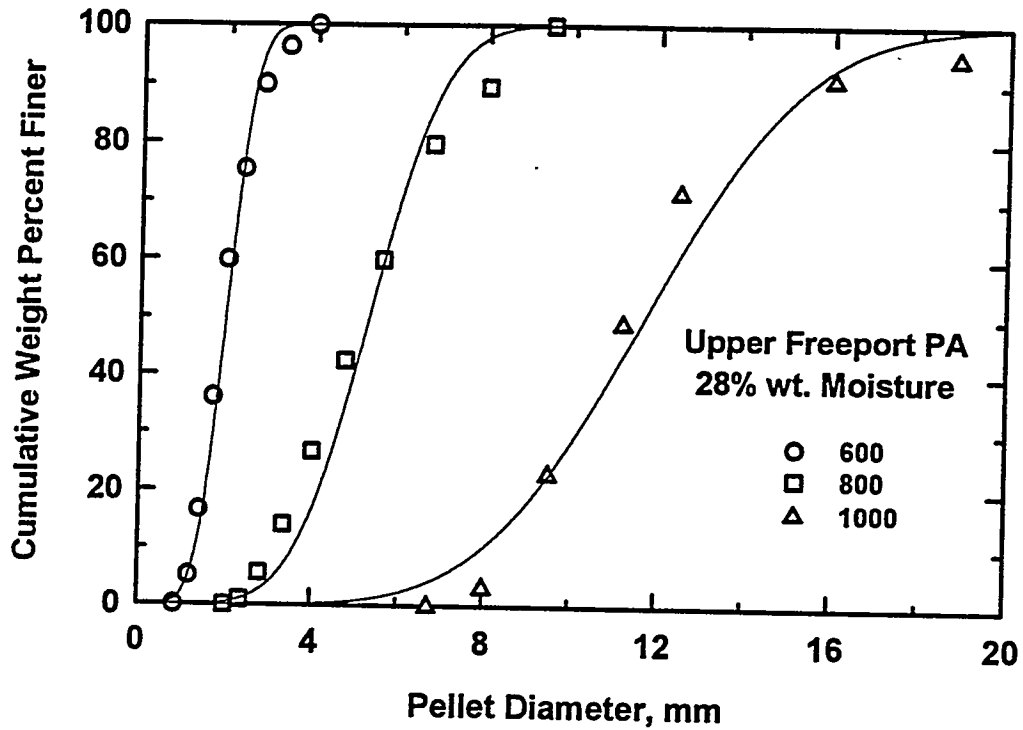
Figure 5.5. Size distribution of Upper Freeport PA (as received) pellets. The solid lines are the predictions of the pellet size distribution by the random coalescence model (Eq. 5.3).
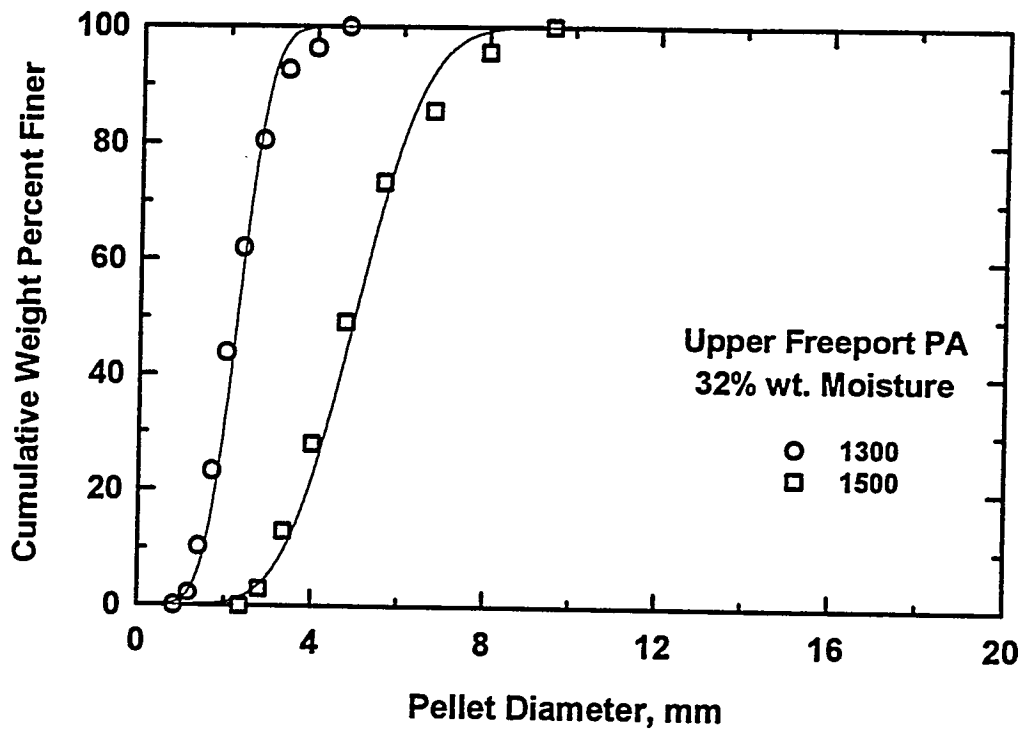
Figure 5.6. Size distributions of pellets produced with weathered Upper Freeport PA coal sample. The size distributions are also in agreement with the random coalesecence model (Eq. 5.3).
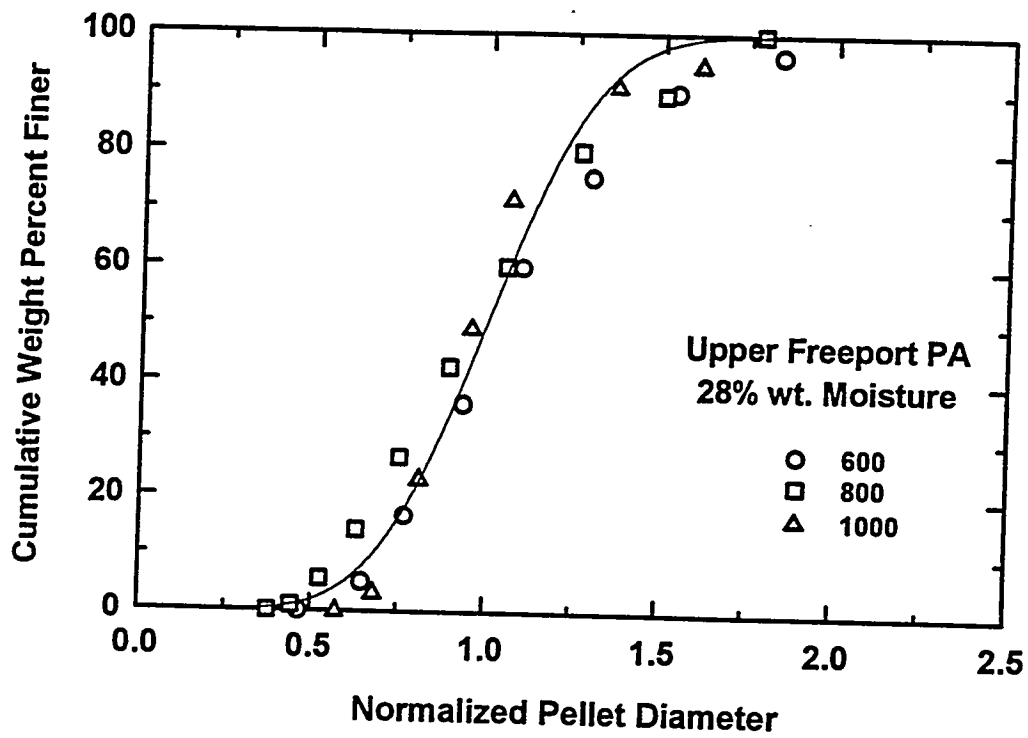
Figure 5.7. Normalized size distributions from the data reported in Figure 5.5 on Upper Freeport PA (as received) sample.
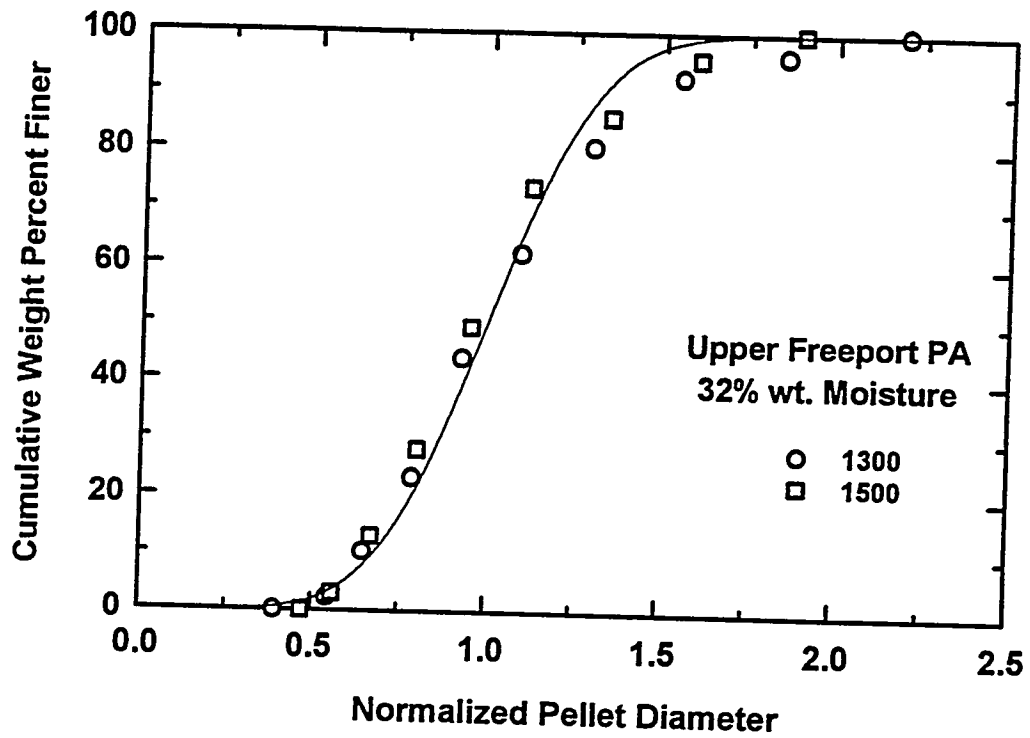
Figure 5.8. Normalized size distributions from the data reported in Figure 5.6 on Upper Freeport PA (weathered) sample.
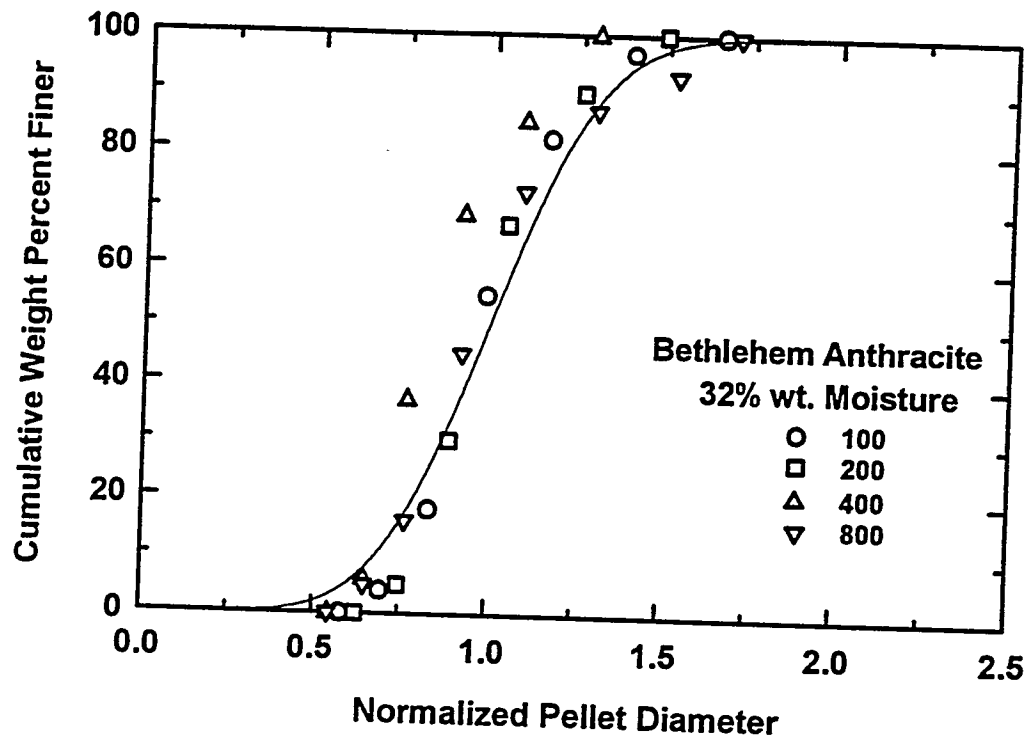
Figure 5.9. Normalized size distributions of the pellets produced with Bethlehem anthracite sample. The figure does not show agreement with the random coalescence model.

5-13
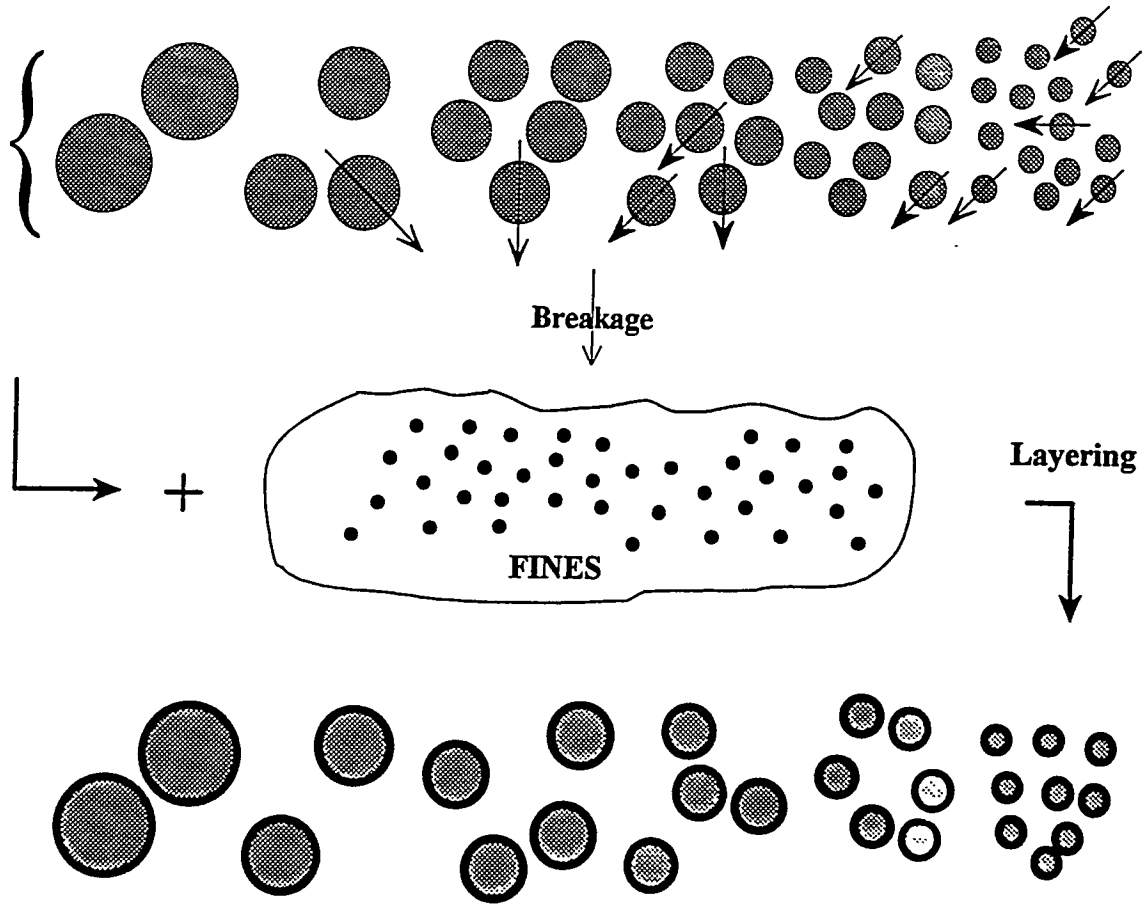
Figure 5.10. Schematic of breakage and layering mechanism.

these assumptions gives rise to the following model equation:

$$\frac{\partial n(\delta,t)}{\partial t} = -\frac{k_B \, n(\delta,t)}{\delta} - \frac{\partial[k_L n(\delta,t)]}{\partial \delta} \tag{5-6}$$

where $n(\delta,t)d\delta$ is the number of agglomerates with pellet diameter in the interval $(\delta,\delta+d\delta)$ and

the symbols $k_B$ and $k_L$ describe the breakage and layering rate parameters respectively and have

the units of mm/sec. A general solution to the above equation does not seem possible.

Nevertheless, it is instructive to carry out a moment analysis of the model equation by making

use of Eq. 2.

$$M_r(t) = \int_0^\infty \delta^r n(\delta,t)d\delta \tag{5-7}$$

From the definition of the moments, we note that $M_3$ corresponds to the total mass of

agglomerating charge in the drum. Further, we recognize that the weight-mean diameter, $\bar{D}_w$

and weight-diameter variance, $\sigma_w^2$ are related to the total moments as follows:

$$\bar{D}_w = \int_0^\infty \delta m(\delta,t)d\delta = \frac{\int_0^\infty \delta \delta^3 n(\delta,t)d\delta}{\delta^3 n(\delta,t)d\delta} = \frac{M_4}{M_3} \tag{5-8}$$

and

$$\sigma_w^2(t) = \frac{M_5}{M_3} - \bar{D}_w^2 \tag{5-9}$$

Multiplying both the right and left hand sides of Eq. 1 by $\delta^r$ and integrate over all $r$, we obtain:

$$\frac{dM_r}{\partial t} = -k_B M_{r-1} + r k_L M_{r-1} = (-k_B + r k_L) M_{r-1} \tag{5-10}$$

for $r \neq 0$.

Since the total mass of the charge remains constant during a batch operation, we require:

$$\frac{dM_3}{dt} = 0 \tag{5-11}$$

Then for the special case of $r = 3$, we have from Eqs. 5 and 6:

$$\frac{dM_3}{dt} = (-k_B + 3k_L) M_2 = 0 \tag{5-12}$$

or

$$k_L = \frac{k_B}{3} \tag{5-13}$$

since $M_2 \neq 0$. This is a very interesting physical requirement that the layering rate constant must always be 1/3 of the breakage rate constant. There is no way, we would have guessed this requirement with out the present mathematical model and analysis.

Thus from Eqs. 8 and 5, we obtain:

$$\frac{dM_r}{\partial t} = \frac{(r-3) k_B M_{r-1}}{3} = (r-3) k_L M_{r-1} \tag{5-14}$$

Further, combining Eqs. 3, 4 and 9 gives rise to:

$$\frac{d\bar{D}_w}{dt} = k_L \tag{5-15}$$

and

$$\frac{d\sigma_w^2}{dt} = 0 \qquad\qquad (5\text{-}16)$$

Solving Eqs. 10 and 11, we get:

$$\bar{D}_w(t) = \bar{D}_w(t_0) + k_L(t-t_0) \qquad\qquad (5\text{-}17)$$

and

$$\sigma_w^2(t) = \sigma_w^2(0) \qquad\qquad (5\text{-}18)$$

where $t_0$ is the time at which the breakage and layering process sets in. These are two other amazingly simple, elegant, but informative results.

Out of all the pelletization kinetic results we obtained, Bethlehem anthracite appears to be governed by the breakage and layering mechanism. Figure 5.11 shows the confirmation of these predictions where the weight-mean diameter, $\bar{D}_w$ is found to increase linearly with time. The constancy requirement for the weight-diameter variance implies that the weight-diameter distribution must undergo a parallel shift as the growth occurs by breakage and layering and the results shown in Figure 5.12 are in reasonable agreement. As a matter of fact, the condition that the variance of the distribution remains constant clearly indicates that the size distribution undergoes a lateral shift with time; the shift occurring due to the smaller agglomerates getting crushed and layered onto the surface of the surviving pellets. Figure 5.13 shows that the size distributions of pellets produced with the Bethlehem anthracite do undergo such a lateral shift.

This is a very impressive mathematical analysis and is in contrast to the breakage and layering model proposed by Capes and Danckwerts (1965), where they assumed that only the
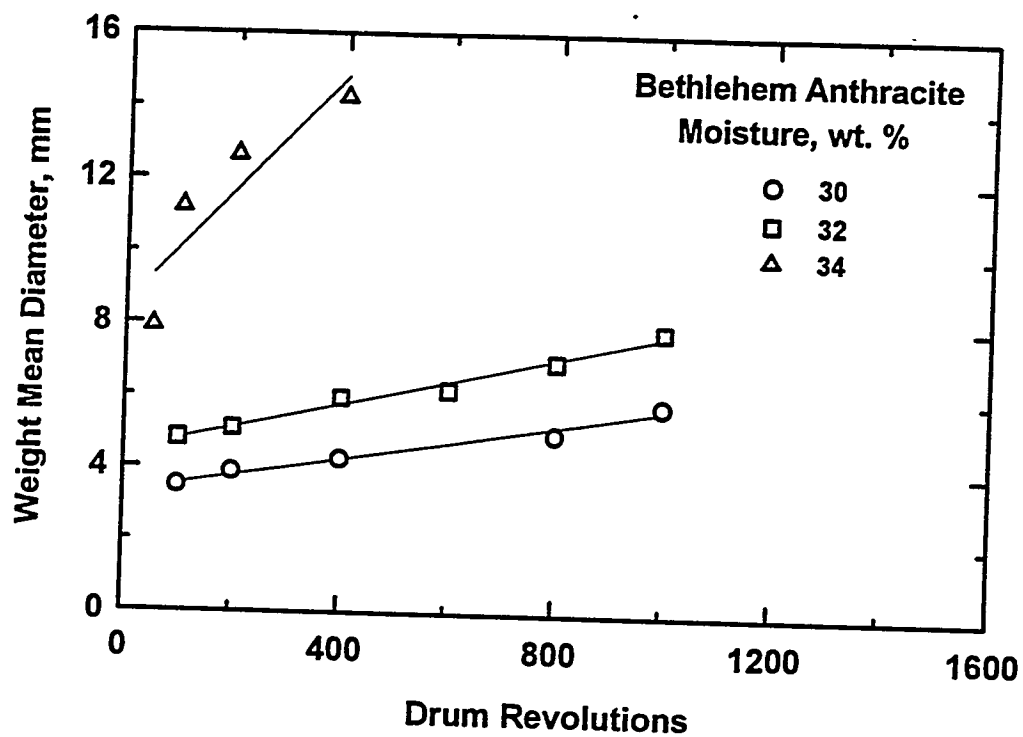
Figure 5.11. Linear growth rate of agglomerates produced with Bethlehem anthracite as predicted by Eq. 5.16.
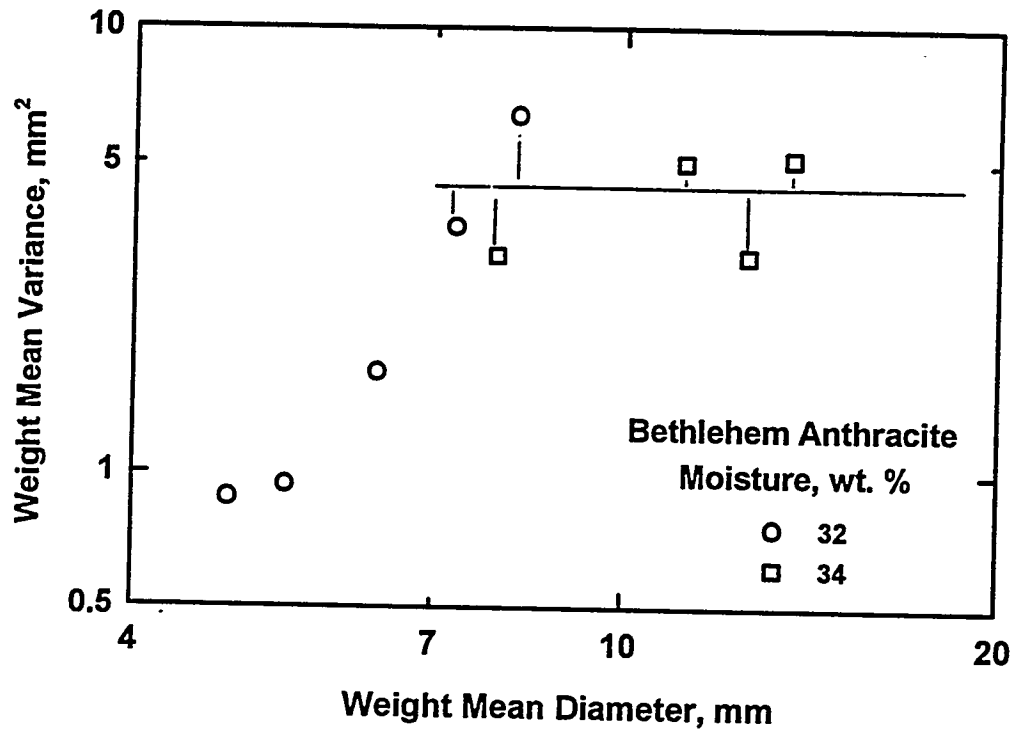
Figure 5.12. Variance versus weight mean diameter of agglomerates produced at 32% and 34% moisture for Bethelehem anthracite.
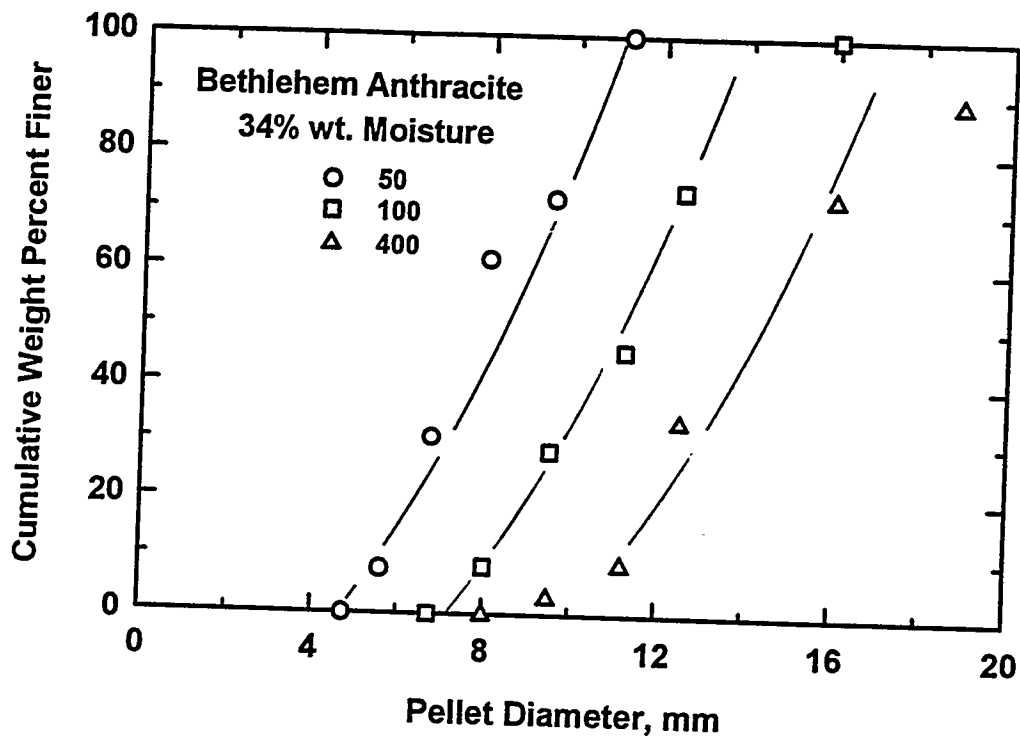
Figure 5.13. Pellet size distributions for 32% moisture run with Behtlehem anthracite.

smallest agglomerates undergoes crushing and the resulting fragments are layered onto the surviving granules. Under this assumption, they have shown that the granule size distribution results in a self-preserving form. A detailed analysis of the granule size distributions (and their moments) obtained with different samples of coal has been made and is shown that there exists no self preserving form. Thus, we conclude that breakage and layering mechanisms during agglomeration of such coals as Bethlehem anthracite occur with finite possibility of granules of all sizes (not just the smallest of the population) subject to breakage and with simultaneous layering of broken fragments on to the surviving agglomerates.

## 6.0 *CoalAgglom*: A DYNAMIC SIMULATOR FOR COAL PELLETIZATION

The unit operation of pelletization is intended to alleviate the problems associated with the handling, storage and processing of fine coals which are being produced in increased quantities. It is expected that there will be three steps in an industrial coal production system [52] -- (i) feed preparation, (ii) pellet production, and (iii) pellet curing. Feed preparation step constitutes mixing of fine coals with desired additives (binders, conditioning agents, fluxes, etc.) and bridging liquids (water, polymeric additives, etc.). The second and main step of agglomerate production is taken place in balling circuits where desired size pellets (commonly known as green pellets) are formed. The final step is where green coal pellets are cured either by thermal drying or simple stockpiling.

It is commonly recognized that the agglomerate production step is fairly critical in that a good quality green pellet is a prerequisite for a good quality cured pellet. Accordingly the pellet production process needs to be better understood and better engineered. Results presented in the earlier sections provide us an increased understanding of the kinetics and modeling of the coal pelletization process. Based on the results of these studies and realizing that a computer simulation system would be quite useful in engineering a coal pelletization circuit, we have undertaken the development of *CoalAgglom*, a simulator for studying the agglomeration of coal pelletizing circuits. This simulator has been adapted from *CCBDrum*, a general purpose simulator that was developed for studying the dynamic behavior of closed circuit balling drums reported earlier [74]. We have incorporated several substantial improvements into *CoalAgglom* which include making the simulator truly object-oriented,

6-1

introducing model correlations that are appropriate for coal agglomeration, and fixing run-time errors that existed in the original versions.

In this section we -- (i) introduce and describe a generalized flowsheet for coal pelletization, (ii) discuss highlights of the mathematical model that form the basis for simulation, (iii) make a broad introduction to *CoalAgglom* simulator, and finally (iv) present a few case studies to demonstrate some of the capabilities of the simulator.

## 6.1 A Generalized Balling Drum Circuit

A conventional balling drum circuit is made up of a closed circuit arrangement of a balling drum and a size separation unit, usually a roller screen (Figure 6.1). Depending on the throughput capacity of the plant there can be several balling circuits running in parallel. Past experiences with the closed-circuit balling circuits indicate that they exhibit a cyclic behavior in the production of finished pellets and in the quantity of undersize agglomerates recycled to the drum [8, 25, 74]. Occasionally, it is found that surging can become so severe that the pelletizing drum or the recycle belt can get overloaded leading to tripping of these units. Only way overload problem can be solved is by manually shoveling and removing the extra material in the drum or the recycle belts. Besides surging is found to result in variation of the green pellet product. Accordingly, new design and operating control strategies are desirable that would help minimize the inherent surging problem. Evaluation of any new strategies in an industrial plant is an expensive proposition especially if the viability is not demonstrated. That is when computer simulation would be found most useful in carrying out a preliminary assessment of any proposed strategies.

The approach that was taken with the *CCBDrum* simulation package was to generalize the balling circuit layout and so as to provide maximum flexibility in its operation [74]. This
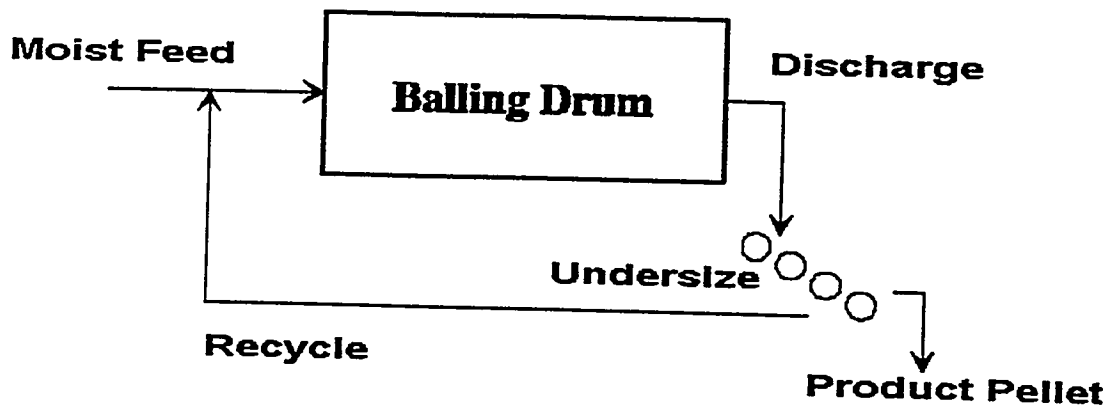
Figure 6.1. Schematic of a conventional closed-circuit pelletizing operation.

involved introducing three additions to the conventional circuit: (i) a feed splitter, (ii) a seeding drum, and (iii) an undersize pellet splitter. The same flowsheet was also adapted for *CoalAgglom*. The generalized flowsheet is shown in Figure 6.2. The first two changes are to enable production of controlled quantity of seed pellets which are then added to the main balling drum. The third change provides the flexibility of recycling only a desired portion but not all of the undersize pellets to the main drum. The portion of recycle that is diverted to stock pile can be brought back to the mixer where it is shredded and fed to the circuit.

This proposed flexible flowsheet could offer a practical solution to improve the performance of the balling circuits and to produce green pellets with consistent quality. Nevertheless, they must be carefully studied in a pilot or a full scale plant and the practical and economic feasibility of such modifications must be established.

## 6.2 Mathematical Model for Simulation

Simulation of the pelletizing circuits requires - (i) relatively straight forward logic and algebraic calculations for combining, splitting and directing the process streams and (ii) a mathematical model for the pelletization process occurring in the seeding drum and the main drum. We have adapted the agglomeration models developed by Sastry [52, 60] which constituting the description of the evolution of the state of the pelletizing charge in time and drum geometry space. The state of the pelletizing charge is described in terms of mass of moist fines and size distribution of the pellets. The changes in the pellet size distribution and mass of moist fines are quantified in terms of the following three agglomeration mechanisms:

- nucleation of moist fines into small seeds,

- coalescence of small seeds resulting in well formed seed pellets, and

- layering of moist fines onto the well formed seed pellets.
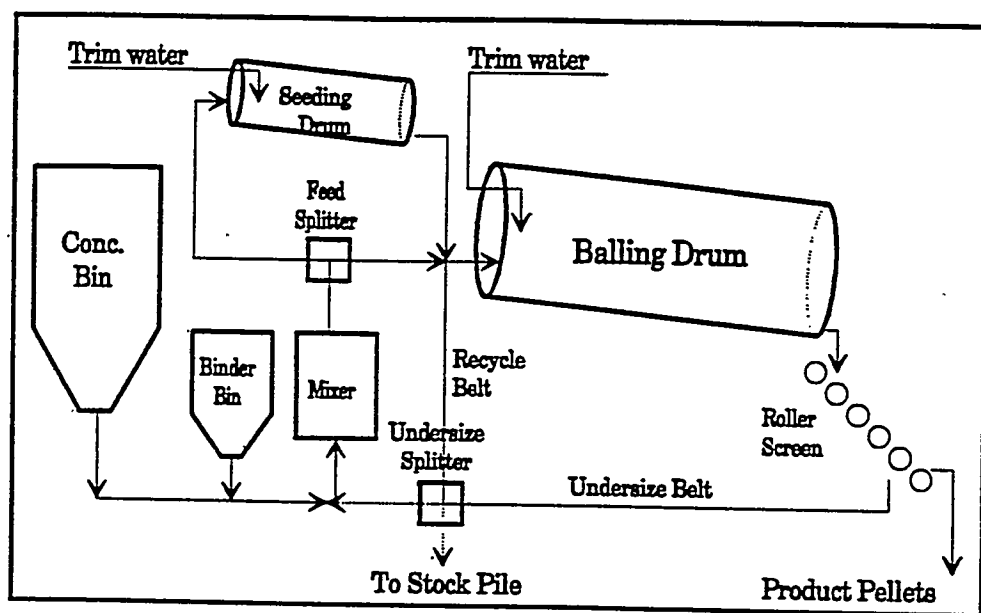
Figure 6.2. Flowsheet of a generalized balling drum circuit that is designed for flexibility of operation.

These three mechanisms are quantified respectively by the process parameters hS, kS and kL. These parameters are determined by the pelletizing nature of the feed material and by such operating conditions as mixer speed, feed moisture, and feed rate.

It is assumed that in the seeding drum the pellet growth occurs only by nucleation and coalescence mechanisms. In the main drum, all the three mechanisms are considered to take place simultaneously but to different extents depending on the process parameters, the amount of recycle (and new seeds from the seeding drum) and feed addition rate to the main drum. Subsequently, size distribution of the discharge pellets from the drum is calculated along with the size distribution and mass flow information on the undersize (recycle) and product pellets.

## 6.3 An Overview of *CoalAgglom*

*CoalAgglom* is developed for IBM/PC-AT and compatible personal computers and runs under Microsoft Windows 3.x with DOS 3.3 (or higher). Minimum recommended hardware requirements are an Intel 80386-25 Mhz processor (with a math coprocessor) with 4 MB of RAM, 80 MB capacity hard disk, a mouse, and an Enhanced Graphics Adapter (EGA) or a Video Graphics Array (VGA) monitor. The software is designed by following advanced object-orientation principles and is written in C++ programming language. The system offers modern graphic-user interface (GUI) consisting of pop-up menus, dialog boxes, along with user interaction through a mouse. As such, the keyboard need be used minimally. Also, such state-of-the-art features as on-line context sensitive help, automatic input data validation, user-definable report generation are incorporated. Following paragraphs present a brief walk through *CoalAgglom* so as to give a touch and feel of this advanced simulator.

To run *CoalAgglom*, one initiates a Windows session and double clicks the *CoalAgglom* icon. After displaying an introductory screen, *CoalAgglom* presents the main menu (Figure 6.3). Each menu item offers a number of choices which are accessed through drop-down submenus shown in Figure 6.3. The first item on the menu is **File**, the subitems offer such file functions as input, save, and print. The **Edit** menu item enables data editing and exchange within *CoalAgglom* system and with other Windows applications. Through the **Inputs** menu, the user can choose to enter the relevant settings, parameters and data to run a *CoalAgglom* simulation. Then by using the **Simulation** menu, the user can either perform a simulation in a batch mode or an interactive mode, or review a previously run simulation. The results can be viewed via the **Outputs** menu in the form of tables and graphs. The **Help** menu item allows access to the help index and version number information of the running application. The help feature is also included in all the *CoalAgglom* screens thus enabling the users to obtain context sensitive help.

For example, when a user selects the **Circuit Settings...** option under the **Inputs** menu item, the window shown in Figure 6.4 would appear. This window presents a fill-in-the-form type data entry dialog box for entering such relevant data about the circuit as dimensions of the seeding drum, the main balling drum. The entire *CoalAgglom* data entry system is provided with automatic data validation. After entering valid data into the **Circuit Settings** screen, the user may want to input the material specifications for the simulation. This can be accomplished either by clicking the **OK** button and entering **Material Specifications...** option through the **Inputs** menu or directly from the **Circuit Settings** screen by clicking **Material> >** button. Either one of the actions takes the simulator to Figure 6.5.
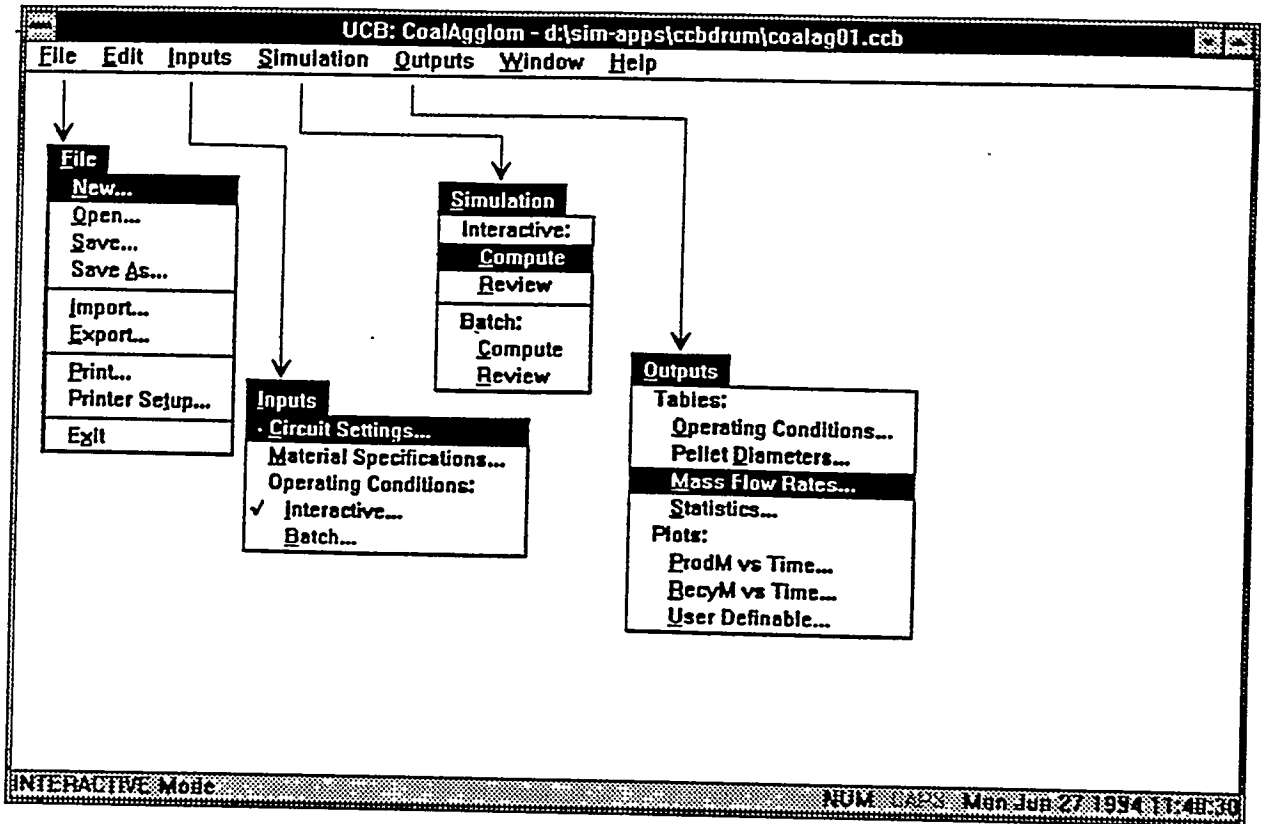
Figure 6.3. The main menu bar and the submenu lists from *CoalAgglom.*
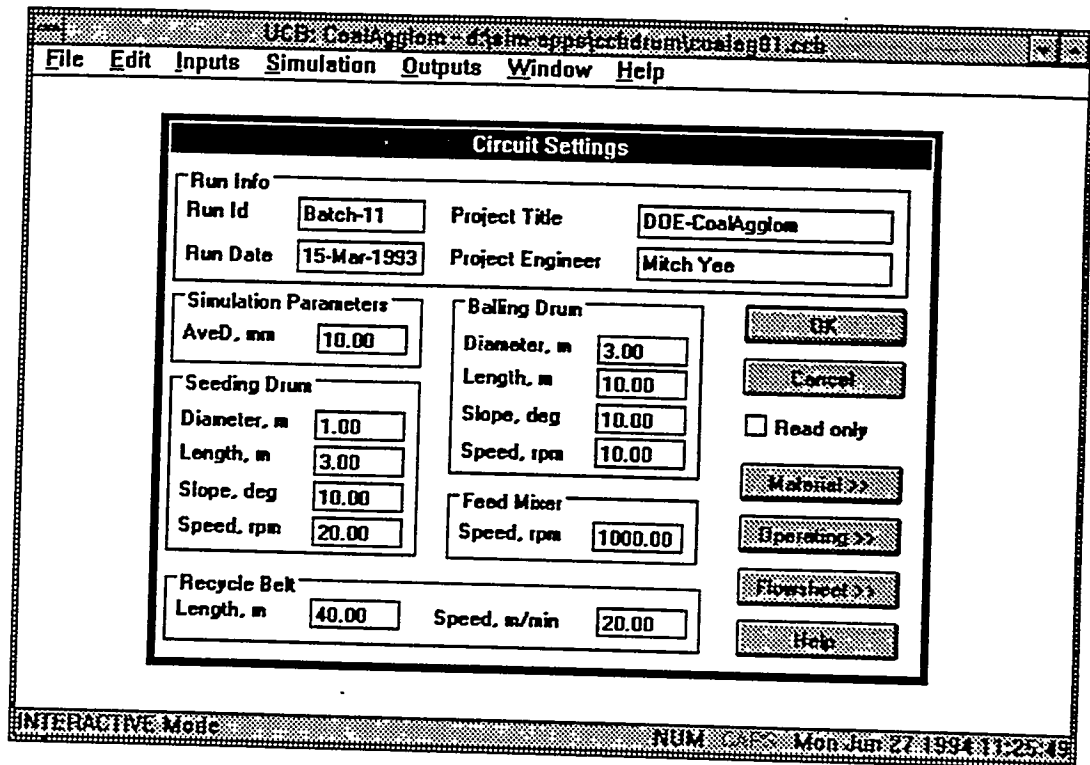
Figure 6.4. Fill-in-the-form type data entry dialog box for **Circuit Settings.**

Figure 6.5 shows an example of the data entry screen for **Material Specifications** in which a range checking error message screen has appeared reporting that the user entered an out of range value of 500 (presumably wanting to enter 5.00) for the specific gravity of the feed material. The user can also obtain context sensitive help by clicking the **Help** button located in each of the *CoalAgglom* screens. Further, the user can obtain the current flowsheet configuration by clicking the **Flowsheet** button located in most of the *CoalAgglom* screens at which time a drop-down box will present the flowsheet shown in Figure 6.2. The user would enter all the necessary inputs and subsequently performs either a **Batch** or **Interactive** simulation. This takes the user to the simulation panel shown in Figure 6.6.

As seen in Figure 6.6, the simulation panel is divided into five subwindows including **Operating Conditions, Simulation Controls,** a table of **Simulation Results,** and two dynamically scrolling plots of simulation results. The simulation run can be controlled by pressing one of the control buttons in the **Simulation Controls** panel: **Go, Restart,** and **End.** The simulation is initiated by clicking the **Go** button which turns into **Pause** which can subsequently clicked to pause the run. At this time the **Pause** button changes to **Continue,** and the user may change the operating conditions such as percent feed to main drum, feed moisture, and screen size in the **Operating Conditions** window either by entering the desired value directly or by clicking the spin buttons. The spin buttons allow the associated value to be incremented or decremented by pressing the up or down arrows respectively. After the desired changes in the operating conditions are made, the user presses the **Continue** button in the **Simulation Controls** subwindow to continue with the simulation. As the simulation is performed the results are displayed in one tabular and two graphical windows with a scroll effect. The simulation is carried out until the preset time limit is reached or when the user
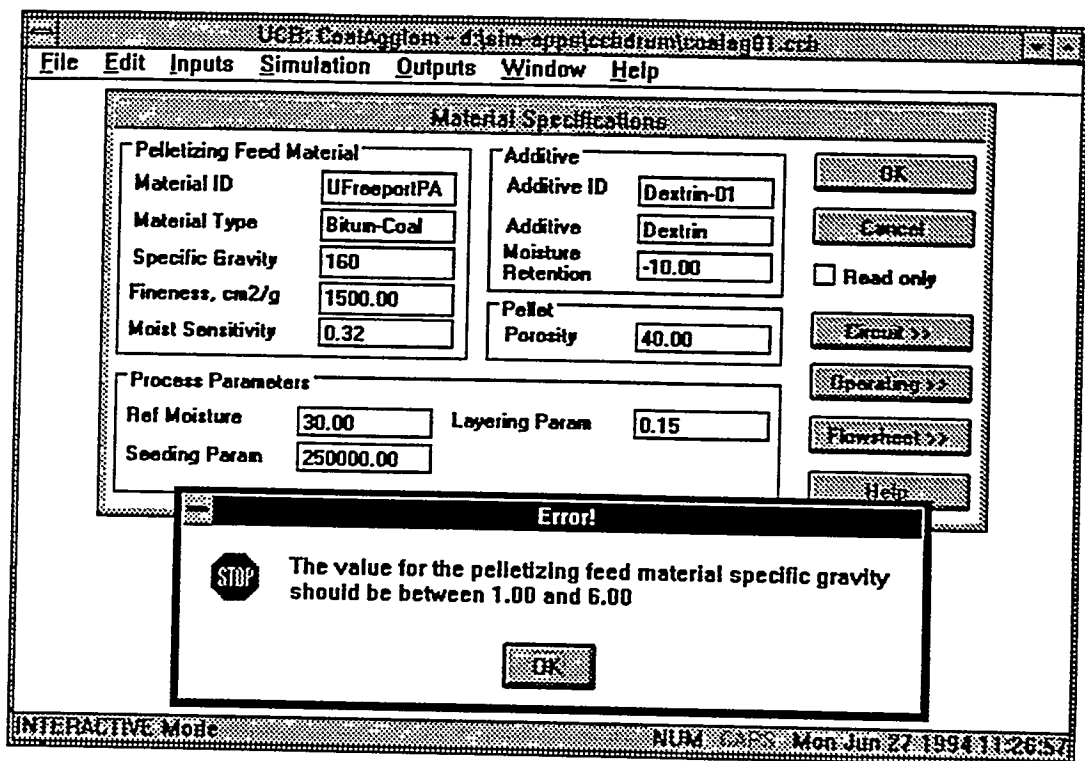
Figure 6.5. The data entry screen for **Material Specifications**. The figure also shows the error message box resulting from range checking on the feed specific gravity.

**UCB: CoalAgglom – d:\sim-apps\ccbdrum\coalag01.ccb**

File   Edit   Inputs   Simulation   Outputs   Window   Help

**Operating Conditions**

| | |
|---|---|
| Circuit feed rate, tph | 30.00 |
| % Feed moisture | 18.50 |
| Additive, kg/t | 0.00 |
| % Feed to SD drum | 0.00 |
| Trim H2O to SD drum, % units | 0.00 |
| Trim H2O to main drum, % | 0.00 |
| % Usize to Recycle | 100.00 |
| Screen opening, mm | 18.00 |

kS  2.53e+005   kL  0.2000

hS  0.1050

**Simulation Review Controls**

Continue | Restart | End | Flowsheet >>

**RecyMass/FeedRate vs. Time**

**ProdMass/FeedRate vs. Time**

**Simulation Results**

| CY# | RecyM | ProdM | ProdD |
|---|---|---|---|
| 47 | 27.12 | 29.38 | 10.75 |
| 48 | 27.74 | 26.70 | 10.72 |
| 49 | 31.04 | 28.60 | 10.70 |
| 50 | 32.44 | 31.70 | 10.72 |
| 51 | 30.74 | 32.03 | 10.73 |
| 52 | 28.71 | 29.85 | 10.74 |
| 53 | 28.87 | 28.39 | 10.73 |
| 54 | 30.49 | 29.21 | 10.72 |
| 55 | 31.28 | 30.78 | 10.72 |
| 56 | 30.50 | 31.04 | 10.73 |
| 57 | 29.46 | 30.00 | 10.73 |
| 58 | 29.47 | 29.22 | 10.73 |

INTERACTIVE Mode                    NUM CAPS  Mon Jun 27 1994 11:48:30

Figure 6.6.  The *CoalAgglom* simulation panel showing the subwindows for operating conditions, simulation controls, and a table and two plots of simulation results.  Plotted results correspond to the response of the balling circuit to a step change in feed moisture.

clicks on **Pause** and **End** buttons in the **Simulation Controls** window. The user can navigate through the **Outputs** menu item to preview and print any one of the output tables and plots.

## 6.4 A Case Study

This section presents simulation results from a few case studies that are intended to demonstrate some of the typical simulation capabilities of *CoalAgglom*. To keep the case studies simple, the runs were made with the circuit settings and material specifications given respectively in Figures 6.4 and 6.5. The operating conditions under which the simulation was performed are listed in Figure 6.7, which is actually a listing produced by the **Outputs/Operating Conditions...** menu option. The simulation was started with a feed rate of 30 tph (all to the main drum) and feed moisture of 30 percent by weight. The simulation time is shown as number of cycles which corresponds to number of residence times in the main drum.

Results from the initial 58 cycles are shown in the two plots given in Figure 6.6. The plots are for the normalized recycle mass which is circulating load to the main drum and for the normalized product pellet mass. It can be seen from Figure 6.6 that by 30 cycles of simulation, the process underwent two surge cycles with circulating load changing between about 180 and 300 percent and production rate fluctuating between 50 and 150 percent of feed addition rate. This surging behavior will continue for ever if the feed moisture is maintained at 30 percent. At 30 cycles, the feed moisture was changed to 32 percent and within about 15 cycles (which correspond to about 8 minutes in the real time) the process became stable with circulating load settling to about 100 percent. This portion of the simulation thus shows that the circuit can be stabilized by properly choosing the feed moisture. The actual level of feed moisture will be determined by such material properties as seeding and layering rate constants.

```
                              Operating Conditions
       Feed  Feed  Addtve  %Feed  -TrimH20%-  Usze  Scrn
 Cy#   Rate  H20%   kg/t    SdDr   Sdng  Main  Recy  Opng
  0   30.00 30.00   0.00   0.00   0.00  0.00 100.0 10.00
*** at time   30 Feed moisture changed to 32.00%
 30   30.00 32.00   0.00   0.00   0.00  0.00 100.0 10.00
*** at time   60 Circuit feed rate changed to 35.00tph
 60   35.00 32.00   0.00   0.00   0.00  0.00 100.0 10.00
*** at time   80 Usize to Recycle changed to 80.00%
 80   35.00 32.00   0.00   0.00   0.00  0.00  80.0 10.00
*** at time   85 Usize to Recycle changed to 85.00%
 85   35.00 32.00   0.00   0.00   0.00  0.00  85.0 10.00
*** at time   90 Usize to Recycle changed to 90.00%
 90   35.00 32.00   0.00   0.00   0.00  0.00  90.0 10.00
*** at time   95 Usize to Recycle changed to 95.00%
 95   35.00 32.00   0.00   0.00   0.00  0.00  95.0 10.00
*** at time  100 Usize to Recycle changed to 100.00%
100   35.00 32.00   0.00   0.00   0.00  0.00 100.0 10.00
*** at time  120 Circuit feed rate changed to 40.00tph
120   40.00 32.00   0.00   0.00   0.00  0.00 100.0 10.00
*** at time  130 Feed to SD drum changed to 10.00%
130   40.00 32.00   0.00  10.00   0.00  0.00 100.0 10.00

 [ Done ]  [ <<Previous ]  [ Next>> ]  [ Print ]      [ Help ]
```

Figure 6.7. Listing of operating conditions that were used for the balling circuit case study.

At cycle time 60, the circuit feed rate was increased to 35 tph (Figure 6.7). The pellet production rate results are shown in Figure 6.8. Again, the circuit becomes cyclic with production rate fluctuating between 60 and 150 percent. A simple manual control strategy of cutting down the feed back of undersize as recycle to the circuit to 80 percent and progressively bringing back to 100 percent within a total of 20 cycles (10 minutes of real time) was tried. Such control strategy is found to bring the system into control as seen in Figure 6.8.

At cycle time 120, circuit feed rate was again increased to 40 tph and the simulator response is shown in Figure 6.9. As expected the system gets back to surging. While manipulating the undersize return to the main drum does work, we wanted to show that tandem operation of a seeding drum and a main drum also works effectively. At cycle time 130, 10 percent of the feed (i.e. 7 tph) was diverted to the seeding drum, and the discharge from the seeding drum is fed to the main drum along with the remaining 90 percent feed (i.e., 63 tph) to the main drum. Within another 20 or so cycles the system again came to stable operation.

The present case study clearly demonstrates that it is possible to stabilize the balling circuits and to increase their throughput by making appropriate changes in the operating conditions.

The fundamental reason for the circuit to become stable is that various strategies (adjusting the feed moisture, manipulating the quantity of undersize pellets returned to the drum as recycle or splitting the feed to generate and introduce fresh seed pellets) that were employed result in controlling the relative occurrence of new seed generation by the mechanisms of nucleation and coalescence and of continued pellet growth by the mechanism of layering. A balance between seed generation and pellet growth must be struck in order that the circuit stays stable.

We must also recognize that there can be some practical limitations in implementing the modifications suggested above. For example, there can be limitation with the mechanical design

**ProdMass/FeedRate vs. Time**
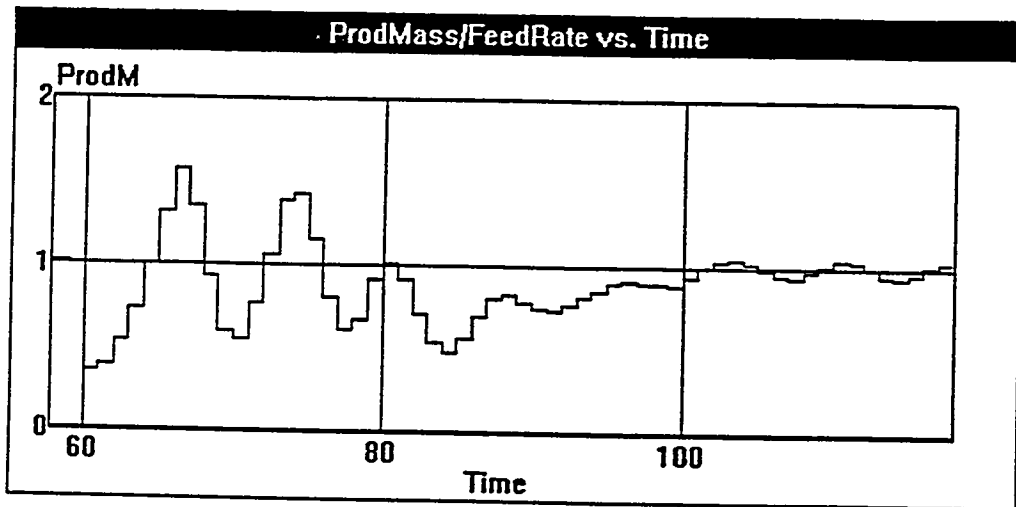
Figure 6.8. Response of the balling circuit - (i) to a step change in feed rate from 30 tph to 50 tph at cycle #60 and (ii) to a manual control action involving amount of undersize fed back as recycle to the circuit so as to bring the circuit to stability.
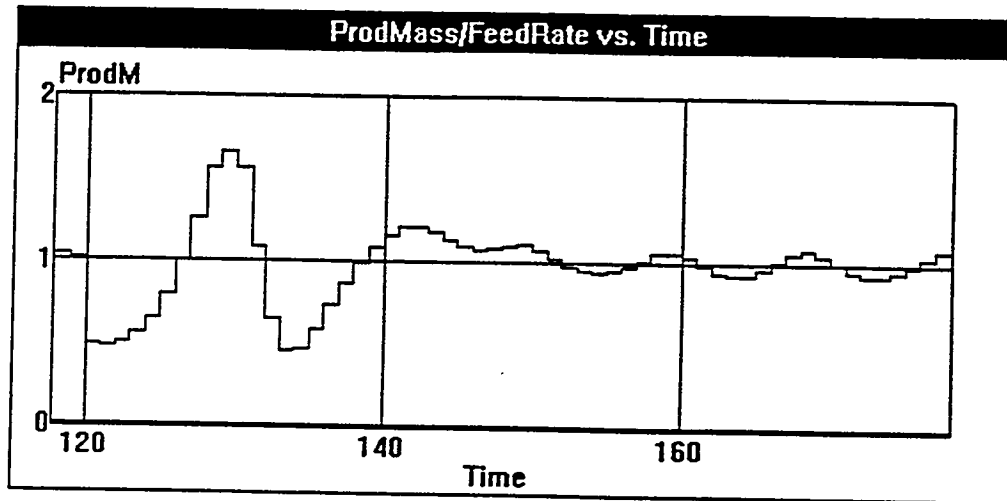
Figure 6.9. Response of the balling circuit - (i) to a step increase in feed rate to 70 tph at cycle #120 and (ii) to the introduction of pre-formed seeds from a seeding drum at cycle #130 to bring the circuit to stability.

and trouble free operation of splitters for circuit feed and undersize pellets. Further, even though the throughput rate can be increased in principle, in practice one must ensure that the load capacities of the drums and the conveyor belts are within operating ranges.

## 6.5 Summary and Conclusions

In this section, we introduced *CoalAgglom*, a dynamic simulator for balling drum circuits. The simulator incorporates state-of-the-art software features including graphic user-interface, automatic data validation and context sensitive help and is easy-to-use. The simulator is based on a mathematical model that was found to adequately describe the behavior of balling circuits. After providing a walk through of the simulator features, a case study to demonstrate the simulation capability of *CoalAgglom* was presented. The case study was intended to demonstrate the flexibility built into the simulator so as to investigate the influence of different operating strategies and the desirability of introducing a seeding drum into the circuit.

It is desirable to incorporate two main features - (i) automatic control system and (ii) online optimization in the future versions of *CoalAgglom* . The first feature would enable offline investigation and identification of new control strategies and the second feature is intended to eventually integrate the *CoalAgglom* simulator with the pelletizing plant control computer system for online optimization of the process.

A complimentary copy of the present version of the simulation program *CoalAgglom* can be obtained by writing to the Principal Investigator at the address shown on the front page.

# 7.0 CONCLUSIONS AND RECOMMENDATIONS

Agglomeration by the unit operation of pelletization consists of tumbling moist fines in drums or discs. Past experimental work and limited commercial practice have shown that pelletization can alleviate the problems associated with fine coals. However, it was recognized that there exists a serious need for delineating the fundamental principles of fine coal pelletization. Accordingly, a research program has been carried involving four specific topics: (i) experimental investigation of coal pelletization kinetics, (ii) understanding the surface principles of coal pelletization, (iii) modeling of coal pelletization processes, and (iv) simulation of fine coal pelletization circuits. This final report summarized the major findings and provided relevant details of this research effort. Following list includes a summary of our research findings and a few recommendations for future studies on coal pelletization.

- Past research on coal pelletization was quite limited and most of the work reported is of applied nature.

- A batch experimental technique has been used to investigate the pelletization of fine coals. Three bituminous coals and one anthracite coal has been studied in this program.

- Reproducibility of the experimental results was found to be a serious problem. Careful experimentation helped improved the reproducibility, but it was often necessary to repeat the experiments several times and use the average results.

- Moisture of coal fines is the most critical variable that determines the rate of pelletization. Also there exists a narrow range of moisture contents over which pelletization is possible. Outside this range, the fine coal feed tends to be too dry or too wet for pelletization.

- Addition of dodecane, a collector for coal flotation, is found to decrease pellet growth. This is attributed to poor efficiency of pellet formation and growth by coalescence due primarily to the increased hydrophobicity with the addition of dodecane. It is desirable to carry out a detailed study in ascertaining the role of collectors on pelletization.

- Addition of dextrin which increases, the wettability of coal, resulted in accelerated rate of pellet growth. It is felt that the structure of coal and such functional groups as carboxylic

and phenolic groups play a major role in determining the influence of dextrin on coal pelletization. A further study should be carried out in understanding these factors.

- Addition of dodecane and dextrin did not influence the pelletization behavior of anthracite. It is anticipated that because anthracite is highly hydrophobic to begin with, addition of surface active agents such as dodecane and dextrin does not result in any noticeable change in its surface properties.

- The three bituminous coals used in the work could be modeled with pellet growth by random coalescence. The process model predicted exponential pellet growth and self-preserving pellet size distributions. Experimental results are found to be in agreement with the model predictions.

- In contrast, growth of anthracite pellets could be best modeled in terms of the occurrence of breakage and layering type growth mechanism. The mathematical model predicted linear pellet growth along with mere shift in the pellet size distributions and the experimental results are in reasonable agreement.

- Qualitative experiments carried out with fine coals showed that raspberry type growth becomes predominant as the coals get finer. The raspberry-shaped coal pellets were found to be fairly weak. It was observed that the raspberry type growth can be reduced by blending coarse coals into the fine coal feeds. A formal quantitative study would be helpful in providing guidelines for industrial operation of coal pelletizing systems.

- Pelletization circuits are known to be inherently unstable in their operation. An interactive simulator called *CoalAgglom* was developed to carry out what-if, why-so and how-to type simulations of coal pelletization process. For example, the simulator can be very useful -- to understand the behavior of pelletization circuits, to identify suitable operating conditions, to investigate novel pelletizing circuit lay-outs and to develop new control strategies for stable operation.

- Detailed process engineering experiments must be carried out in the future with a view to validate the mathematical models and to quantify such mechanisms of pellet formation and growth as nucleation, coalescence, layering, and breakage. These studies would then enable to quantitatively describe a given fine coal sample, to perform computer simulations that are most relevant to the fine coal under investigation and eventually to provide the necessary guidelines for the optimal design and efficient operation of coal pelletization circuits.

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