

A LABORATORY STUDY OF
AGGLOMERATION IN COAL GASIFICATION

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INTRODUCTION

The agglomerating tendencies of coal during gasification, especially during various stages of reaction, is a complex reaction.

Several investigators have studied the behavior of carbon-free ash. Stallmann and Neavel (1) found that the agglomerates of fused ash appear to form at temperatures below the initial deformation temperature, as defined by the ASTM fusibility test (2). Huffman, et al. (3) observed that significant particle melting of the ashes occurred at temperatures as much as 200-400°C below the ASTM initial deformation temperature. They also noted that melting was greatly accelerated under reducing conditions. Rehmat and Saxena (4) argued that it is not necessary for every reacting particle to attain the ash fusion temperature in order for agglomeration to occur. Only a few particles are required to reach the ash melting temperature. These particles form the nuclei for the formation of the ash agglomerates, and this process will indeed consume the ash produced from other particles.

In a fluidized bed, Langston and Stephens (5) recognized that the defluidizing tendency or the stickiness of the particles is directly proportional to the area of contact, the adhesive property of the particles, and inversely proportional to their momentum. Mason and Patel (6) felt that agglomeration depends on fluidized bed temperature, bed ash concentration, average particle size, superficial velocity, and bed height. Goldberger (7) and Siegell (8) both found that the ash softening temperature had no effect on the defluidization characterization for ash samples in their experiments. Basu (9) concluded that defluidization of a fluid bed is governed by the sintering characteristics of the bed material.

In some gasification processes, however, the carbon is only partially gasified in a reducing atmosphere and the remaining carbon is oxidized in a separate combustion zone (10). In addition, an inert solid is present which serves as a heat carrier and a diluent for the coal. The agglomerating tendencies of coal in such a process are different from the behavior of carbon-free ash.

In this paper, we have evaluated the agglomeration of coal, char, and ash during various stages of reaction. This included a range of temperatures, under reducing and oxidizing conditions, and in a mixture with an inert solid.

EXPERIMENTAL

Sample Preparation - Table I shows the proximate and the ultimate analyses of the three subject coals. Table II summarizes the mineral analyses of the ashes which were generated by heating the

coals in air to 538-649°C and screening all samples to pass between 100-mesh and 200-mesh U.S. standard screen. When sand was used, it was 16-30 mesh.

TABLE I
COAL ANALYSIS

	North Dakota <u>Lignite</u>	Western <u>Subbituminous</u>	
		<u>No. 1</u>	<u>No. 2</u>
Proximate Analysis, %			
Moisture	12.3	11.3	8.0
Volatile	34.5	31.3	13.0
Fixed Carbon	44.2	40.2	60.0
Ash	9.1	17.2	19.0
Ultimate Analysis, %			
H ₂ O	12.3	11.3	8.0
C	60.2	54.8	64.9
H	4.2	4.1	2.0
N	0.9	1.1	0.9
S	1.0	0.8	2.6
O	12.3	10.8	2.5
Ash	9.1	17.2	19.0

TABLE II
MINERAL ANALYSIS OF ASH

<u>% of Ash</u>	North Dakota <u>Lignite</u>	Western <u>Subbituminous</u>	
		<u>No. 1</u>	<u>No. 2</u>
SiO ₂	39.8	59.3	23.8
Al ₂ O ₃	16.4	25.7	9.2
TiO ₂	0.8	0.9	0.5
Fe ₂ O ₃	8.1	5.2	14.3
CaO	13.0	2.7	14.2
MgO	3.9	1.0	4.5
K ₂ O	0.2	0.8	0.5
Na ₂ O	0.8	1.9	5.9
SO ₃	15.9	2.2	26.6
P ₂ O ₅	0.5	0.2	0.2
SrO	0.4	0.0	0.1
BaO	0.1	0.1	0.3
Mn ₃ O ₄	0.2	0.0	0.1

Equipment and Procedures - Figure 1 shows the experimental setup employed. In a typical experiment, the furnace was purged with nitrogen and preheated to 538°C before a 0.5-g sample of material was placed in a ceramic boat and positioned in the center of the furnace. The sample was heated to the desired temperature in nitrogen before either a reducing or an oxidizing gas was passed over the sample for a predetermined time. The gas flow velocity was approximately 0.8 cm/sec. After each experiment, the sample was allowed to cool outside the furnace and sieved to determine the extent of agglomeration. The extent of agglomeration is defined as the weight percent retained on a mechanically vibrated 100-mesh screen.

The oxidizing gas was air. The reducing gas consisted of 51.8 vol % H₂, 25.8 vol % CO, 18.8 vol % H₂O, 1.7 vol % H₂S, 1.1 vol % CO₂, and 0.8 vol % NH₃. In some experiments, water was not used.

The average mass balance varied from a low of 96.9% for the North Dakota Lignite ash to a high of 100.8% for the Western Subbituminous No. 1, assuming complete decomposition of the sulfate and phosphate from the ash under experimental conditions.

In either a reducing or an oxidizing atmosphere at a given temperature, run lengths between 10 min. and 30 min. caused no significant difference in agglomeration, indicating that the agglomeration is complete by 10 min. Varying the sample size from 0.5-1.5 g showed no significant difference in the results, indicating that there is no particle mass transfer limitation in our tests.

RESULTS

Ash Agglomeration - Figure 2 shows the results with carbon-free ashes in the reducing gas. At 927°C, there is no significant agglomeration of any of the samples. The Western Subbituminous No. 1 sample developed substantial agglomeration at 982-1038°C, the No. 2 sample at 1038-1093°C, and the North Dakota Lignite ash at 1093-1149°C. All three samples were completely agglomerated at 1149°C. Figure 3 shows that in a reducing environment, the agglomerates transformed from a loosely packed ash at low temperature to a glassy melt at high temperature.

Figure 4 shows the effects of both reducing and oxidizing environments at 1149°C. Both conditions produce agglomeration, but at this temperature the reducing gas leads to more pronounced agglomeration. There was no agglomeration in a nitrogen environment. Thus, agglomeration is related to reactions between components in the ash and in the surrounding gas environment.

Effect of Carbon Conversion - Western Subbituminous Coal No. 1 was used to prepare samples of varying carbon content. The samples were tested in a variety of environments. Figure 5 shows that the agglomeration tendency correlates well with carbon content. No agglomeration occurs until more than 80% of the carbon is removed. Thus, modest amounts of carbon will retard ash agglomeration, even at quite severe conditions. Figure 6 shows that at 64%

carbon conversion, the sample looks much like the starting coal. At 99% conversion, the sample has agglomerated some, but still not to the extent as a carbon-free ash.

Effect of Inert Solids - Since carbon-free ash samples agglomerated the most, we mixed them with silica sand at a 1/10 ratio. Table III shows that agglomeration decreased in both reducing and oxidizing environments. Figure 7 shows that this decrease in agglomeration is quite dramatic in the reducing atmosphere. The ash samples agglomerated and shrunk; on the other hand, the particles in the ash/sand mixtures are well separated and bulky.

TABLE III
AGGLOMERATION OF
WESTERN SUBBITUMINOUS NO. 1

	% Agglomeration		
	1038°C	1093°C	1149°C
Ash Only			
N ₂	0	0	0
Reducing, 10 Min.	74	97	100
Oxidizing, 10 Min.	78	98	100
Oxidizing, 30 Min.	74	99	100
Ash + Sand			
N ₂	1	1	2
Reducing, 10 Min.	64	94	91
Reducing, 30 Min.	65	86	91
Oxidizing, 30 Min.	13	60	87

DISCUSSION

Agglomeration Temperature - Stallmann and Neavel (2) defined the agglomeration temperature as the 50% point on a curve like those in Figure 2, which corresponds to the steep part of these curves. Figure 8 compares the agglomeration temperature from both our experiments and those of Stallmann and Neavel (2) with the ASTM initial deformation temperature. This figure confirms the observations made by Stallmann and Neavel that the agglomeration temperature is a few hundred degrees lower than the ASTM temperature and that the ASTM initial deformation temperature cannot be correlated to the agglomeration temperature.

Figure 9 indicates that the agglomeration temperature for these three coal ashes decreases linearly with increasing sodium content of the ash. No other ash component showed a consistent relationship to agglomeration.

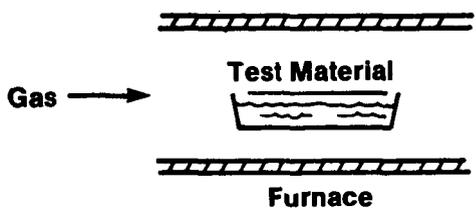
Process Implications - The control of agglomeration is important in the design and operation of a coal gasification or combustion process. We found that agglomeration depends on both coal

properties and on process conditions. In particular, the agglomeration of partially converted coal is negligible up to 80 wt % conversion. At higher conversions, agglomeration could be controlled by diluting the reacting material with inert materials such as sand. This study also indicated that agglomeration occurs on a shorter time scale than the nominal residence time in all but the most kinetically rapid gasification systems. These observations suggest that a gasification process which requires only partial gasification in a reducing atmosphere and which employs a diluent inert, such as the one disclosed by Mitchell, et al. (10), can greatly reduce the agglomeration problem which may occur in processing coal.

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FIGURE 1
ASH AGGLOMERATION TEST



Initial Material
100-200 Mesh

Agglomeration
Material Retained on a 100 Mesh Screen
After Test Expressed as Wt % Ash

FIGURE 2
ASH AGGLOMERATION IN REDUCING GAS

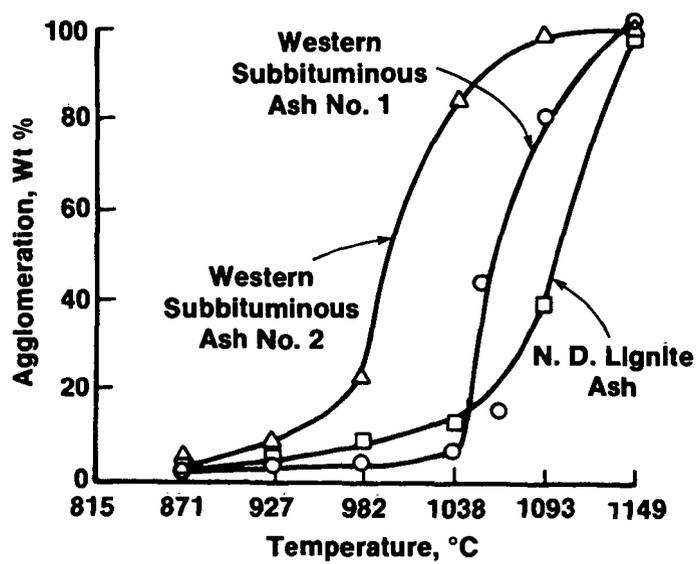


FIGURE 3
EFFECT OF TEMPERATURE ON
AGGLOMERATION



1038°C, Reducing Gas

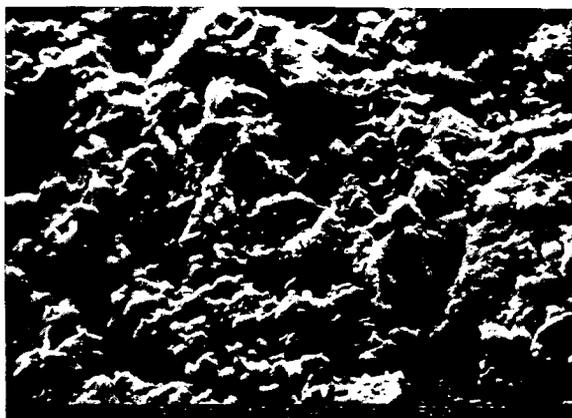


1149°C, Reducing Gas

FIGURE 4
EFFECT OF GAS ENVIRONMENT ON
AGGLOMERATION



Reducing Gas at 1149°C



Oxidizing Gas at 1149°C

FIGURE 5

ASH AGGLOMERATION USING A WESTERN SUBBITUMINOUS COAL

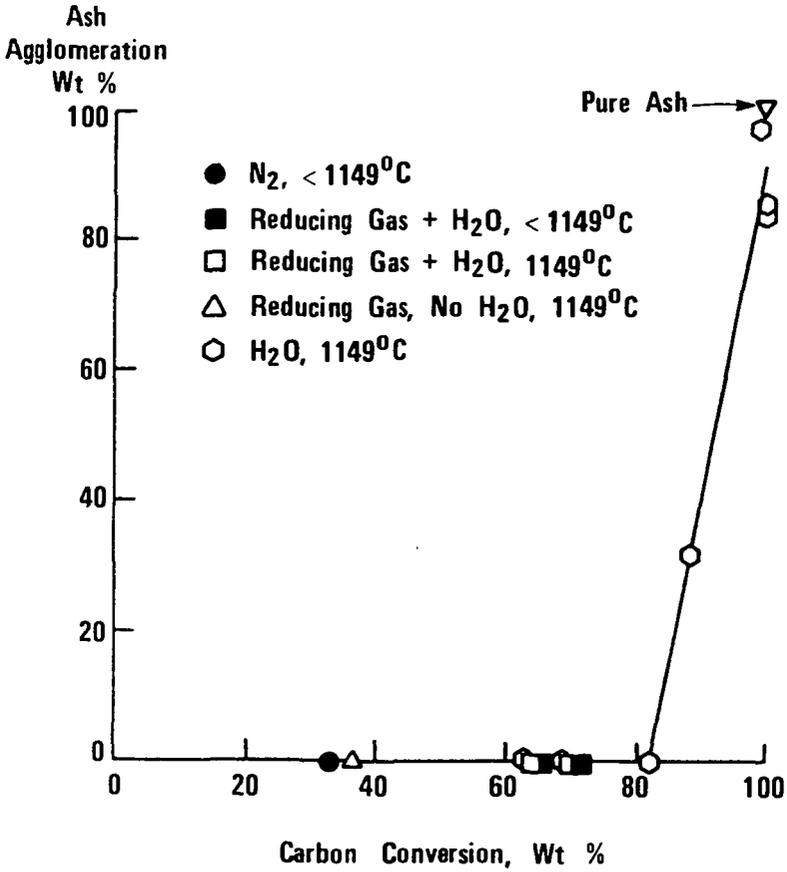
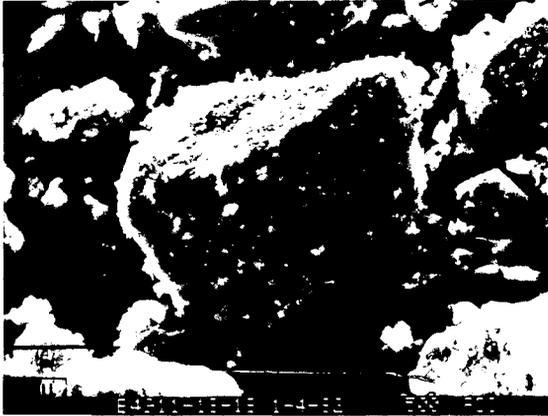


FIGURE 6
AGGLOMERATION OF
WESTERN SUBBITUMINOUS COAL NO. 2 AT 1149°C



Reducing Gas, 64 Wt % Conversion



Steam, 99 Wt % Conversion

FIGURE 7
EFFECT OF TEMPERATURE AND
INERT MATERIAL ON
ASH AGGLOMERATION IN REDUCING GAS

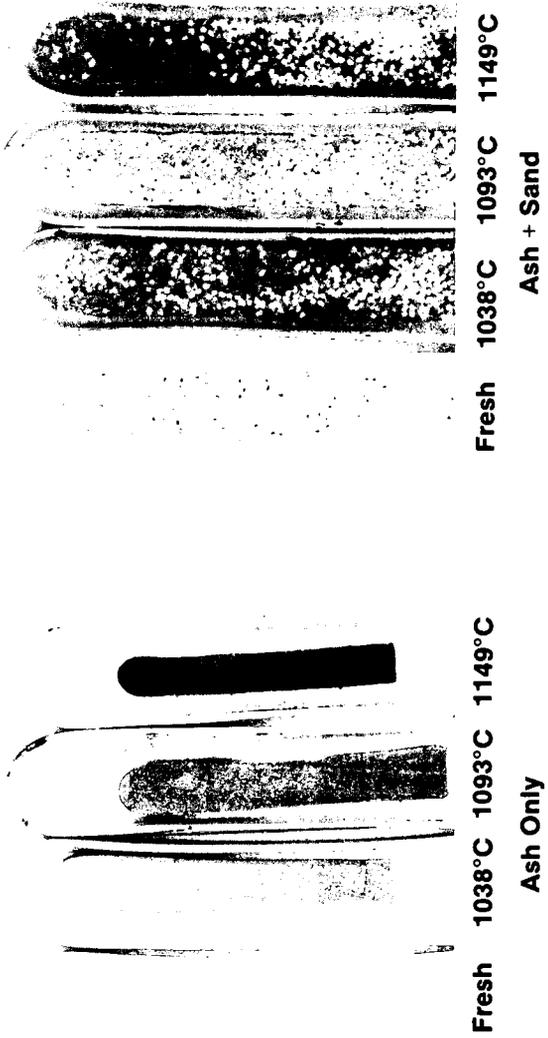


FIGURE 8
AGGLOMERATION TEMPERATURE VERSUS
INITIAL DEFORMATION TEMPERATURE

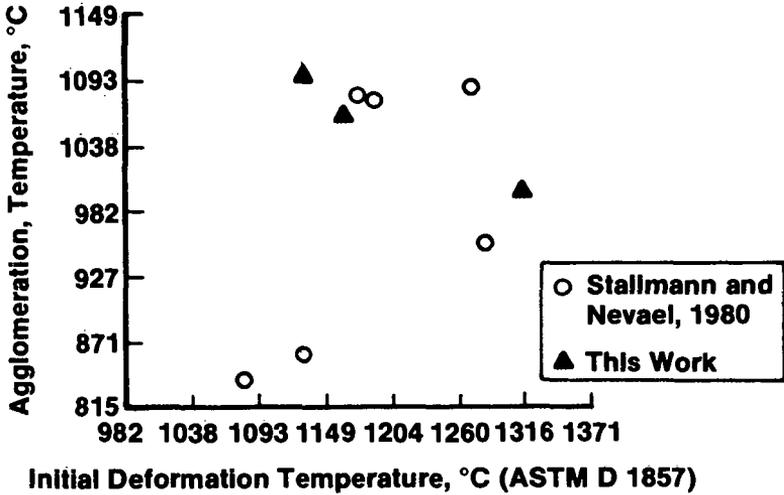


FIGURE 9
THE EFFECT OF SODIUM CONTENT ON
ASH AGGLOMERATION

