

# **PRODUCTION OF MANUFACTURED AGGREGATES FROM FLUE GAS DESULFURIZATION BY-PRODUCTS**

M. M. Wu, D.C. McCoy, M. L. Fenger, R. O. Scandrol, R. A. Winschel,  
J. A. Withum and R. M. Statnick

## **CONSOL Inc.**

Research & Development  
4000 Brownsville Road  
Library, PA 15129

## **ABSTRACT**

CONSOL R&D has developed a disk pelletization process to produce manufactured aggregates from the by-products of various technologies designed to reduce sulfur emissions produced from coal utilization. Aggregates have been produced from the by-products of the Coolside and LIMB sorbent injection, the fluidized-bed combustion (FBC), spray dryer absorption (SDA), and lime and limestone wet flue gas desulfurization (FGD) processes. The aggregates produced meet the general specifications for use as road aggregate in road construction and for use as lightweight aggregate in concrete masonry units. Small field demonstrations with 1200 lb to 5000 lb of manufactured aggregates were conducted using aggregates produced from FBC ash and lime wet FGD sludge in road construction and using aggregates made from SDA ash and lime wet FGD sludge to manufacture concrete blocks. The aggregates for this work were produced with a bench-scale (200-400 lb batch) unit.

In 1999, CONSOL R&D constructed and operated a 500 lb/hr integrated, continuous pilot plant. A variety of aggregate products were produced from lime wet FGD sludge. The pilot plant test successfully demonstrated the continuous, integrated operation of the process. The pilot plant demonstration was a major step toward commercialization of manufactured aggregate production from FGD by-products.

In this paper, progress made in the production of aggregates from dry FGD (Coolside, LIMB, SDA) and FBC by-products, and lime wet FGD sludge is discussed. The discussion covers bench-scale and pilot plant aggregate production and aggregate field demonstrations.

## **INTRODUCTION**

The production of manufactured aggregates provides a potentially significant opportunity to utilize coal combustion by-products. Construction aggregates, traditionally produced from natural stone and sand, constitute about 80% by volume of the concrete used in structural materials and road construction. The U.S. consumption of natural aggregates is about two billion tons per year. The replacement of natural aggregates with aggregates manufactured from coal combustion by-products could provide an economical high-volume use and substantially expand markets for combustion by-products. Many other utilization options (e.g., structural fill) are limited by consumption volume, seasonal demands, and problems in handling, transportation, and storage.

Over the last ten years, CONSOL R&D developed a disk pelletization process to produce manufactured aggregates from coal combustion by-products. This process initially was developed to produce construction aggregates from dry FGD (Coolside, LIMB, SDA) and FBC by-products. In 1996, the effort was redirected to produce construction aggregates from wet

FGD sludge (lime and limestone processes). The aggregates were made in a three-step process involving mixing, disk pelletization, and curing. The properties of the manufactured aggregates were characterized and compared with specifications for use in road and structural construction. The aggregate product may be used as produced, or it may be crushed to meet a specific size classification or to produce angular pieces for use in paving. A variety of useful products, such as lightweight aggregates, American Association of State Highway Transportation Officials (AASHTO) Class A road aggregates, and concrete aggregates was produced, depending on the specific feedstock materials and the operation of the pelletization process. The Ohio Coal Development Office (OCDO) provided partial financial support for bench-scale aggregate production from dry FGD materials (Coolside, LIMB and FBC) and lime wet FGD sludge and aggregate field demonstration. Other project participants are CONSOL R&D, Trumbull Corp., Garick Corp., and Walden Industries Inc. In 1999, a pilot plant was constructed and operated to demonstrate continuous, fully integrated operation of the process with financial support from the U. S. DOE, Duquesne Light Co., SynAggs, Inc. and CONSOL R&D.

## **RESULTS AND DISCUSSION**

### **Coolside and LIMB Ash Aggregates**

The initial objective of CONSOL's aggregate production work was to reduce the waste management costs and the total levelized process cost of two clean coal technologies: Coolside and LIMB sorbent injection FGD processes. The Coolside and LIMB processes were demonstrated successfully at the Ohio Edison Edgewater Station in Lorain, OH, from 1989 to 1992 as part of the DOE Clean Coal Technology (CCT) program.<sup>1</sup> These two processes produce a dry solid waste that must be managed to ensure environmental compliance and economic feasibility. High-strength manufactured aggregates with low abrasion indices were made from Coolside and LIMB ashes. Both aggregates met the ASTM C-331 specifications for lightweight aggregates for concrete masonry units.<sup>2</sup> However, the poor market penetration of the Coolside and LIMB processes did not justify the further development of aggregate production from Coolside and LIMB ashes.

### **FBC Ash Aggregate**

FBC is a commercial technology that combines steam generation with SO<sub>2</sub> control. FBC ash is generated at a temperature of about 1600 °F during coal combustion. The dry ash consists mainly of calcium sulfate, calcium oxide, and coal ash. The manufactured aggregates produced from the dry FBC ash meet the AASHTO Class A aggregate specifications. Although not required by specification, additional tests were conducted to evaluate the effects of natural weathering and freeze/thaw on aggregate properties. The aggregates degraded when they were fully immersed in water during long-term natural weathering and laboratory freeze/thaw tests. Durability was improved by blending FBC ash with pulverized coal (p.c.) fly ash.<sup>3</sup> Approximately 3000 lb of FBC ash aggregates meeting AASHTO Class A aggregate specifications (Table 1) were produced in a semi-continuous batch operation. The raw materials were FBC ash, p.c. fly ash, and water. Since the aggregates produced were spherical, 70 wt % was rough crushed to produce angular pieces for improved load bearing. Marshall stability testing indicated that the aggregates produced an excellent asphalt mix. Results of the Marshall stability tests using various asphalt mix designs are shown in Table 2.

The FBC ash aggregates were used as approximately 50 vol % (43 wt %) of the coarse aggregates in a flexible bituminous (FB) asphalt mix. The other components of the asphalt mix included commercial No. 8 crushed limestone and about 6.5% AC-20 asphalt. No fine aggregate was used. A 12' x 35' x 1.5-2" test patch was laid as a surface course on a high-volume truck road in West Mifflin, PA, in April 1995. The test patch was monitored for over a year by visual

inspection and by collecting and characterizing core samples on a monthly basis from April 28, 1995, to May 30, 1996. FB mixes are designed to be relatively permeable to water. The heavy truck traffic and the permeability of the paving mix made this a severe test of the aggregate durability.

Visual inspections showed that the pavement surface remained uniformly hard and revealed no degradation of the pavement or the aggregate. The surface became too hard to obtain intact core samples with the existing equipment after December 1995. New coring equipment allowed representative core samples to be collected on May 30, 1996. The core samples were extracted with tetrahydrofuran (THF) to remove the asphalt, then screened to determine the size distribution of the aggregates (Table 3). The near-constant size distribution of the core samples indicates that the aggregates maintained their integrity during use for one year. No fines (-8 mesh) were present in the original aggregate, although 33 wt % to 40 wt % fines existed in the recovered cores. The fines recovered from the cores could have been due to extraneous material, degradation of either the natural or manufactured aggregates, or they could have been generated during the coring operation. To determine the source of the fines (extraneous material, or degraded FBC ash aggregate or limestone), the 8 x 50 mesh and -50 mesh fractions of the THF-extracted core samples were analyzed. The source of the fines in the recovered core samples was estimated from elemental analyses, as follows. The FBC ash aggregate content was estimated from the sulfur content, assuming that the sulfur is derived entirely from FBC ash aggregates. The  $\text{CaCO}_3$  content, which represents the limestone, was estimated from the difference between the total Ca in each size fraction and the Ca from the FBC ash aggregates in that fraction. The remaining material not categorized as either FBC ash aggregates or  $\text{CaCO}_3$  was categorized as extraneous material, which includes impurities in the limestone aggregate, and other stones, sand, and dirt deposited in the test patch by traffic. Table 3 lists the estimated sources of the recovered fines. Elemental analysis indicated that the extraneous matter had an  $\text{SiO}_2$  content of ca. 50%; thus, it probably represents sand and dirt. The ratio of FBC ash aggregate to limestone aggregate in the fine fractions remained nearly constant throughout the test period (Table 3). It is concluded that the fines in the cores arose from extraneous materials and from aggregate breakage during the coring operation. All evidence indicates that the FBC ash aggregates were as durable as the limestone.<sup>4,5</sup>

### **Spray Dryer Ash Aggregate**

The spray dryer adsorption process is a commercial technology that removes  $\text{SO}_2$  from the flue gas downstream of the air preheater at a temperature below 300 °F. The dry ash mainly consists of calcium sulfite, calcium hydroxide, and coal fly ash. The manufactured aggregates produced from spray dryer ash meet the ASTM C 331 lightweight aggregate specifications. Both long-term natural weathering and laboratory freeze/thaw tests indicate that spray dryer ash aggregate has good durability.<sup>2</sup>

In 1995 and 1996, two batches (1200 lb and 1700 lb, respectively) of lightweight aggregates were produced from spray dryer FGD by-product. The raw materials consisted of spray dryer ash, lime, and water. The properties of the resulting aggregates are shown in Table 4.

The spray dryer ash aggregates were used to produce two batches of lightweight concrete blocks in a commercial block plant in Ohio. The 1995 batch of aggregates was used to produce solid-core 6" x 8" x 16" blocks. The 1996 batch of aggregates was used to produce 75% solid, two-hollow-core, 6" x 8" x 16" concrete blocks. The properties of the blocks are shown in Table 5. Both batches of blocks met ASTM C-90 specifications in unit weight, water absorption and compressive strength for use in concrete masonry units.

### **FGD Sludge Aggregates**

Although CONSOL's manufactured aggregate development program was technically successful for FBC and spray dryer by-products, neither the FBC process nor the spray dryer FGD process is as widely deployed on coal-fired boilers in the U.S. as wet FGD scrubbing using lime and limestone processes. The sludge mainly consists of calcium sulfite, calcium sulfate or a mixture of both and associated water, depending on scrubber types and operation conditions. About 25 MM tons of unfixated FGD sludge (dry basis) is produced in the U.S., yet only 9% is utilized.<sup>6</sup> Therefore, research to produce manufactured aggregates from lime and limestone wet FGD sludge for use in road and structural construction was initiated in 1996. OCDO supported the bench-scale (200-400 lb batch) study of aggregate production from lime wet FGD sludge and the field demonstration of FGD sludge aggregate in road construction and concrete block manufacture. In 1999, CONSOL co-funded, constructed and operated a 500 lb/hr pilot plant to demonstrate continuous, fully integrated operation of the process. Additional financial support was provided by U. S. DOE, Duquesne Light Company and SynAggs, Inc. The progress of the OCDO supported bench-scale aggregate production and field demonstration, and DOE-supported pilot plant demonstration is summarized below.

### Bench-scale Aggregate Production and Field Demonstration

Road Construction. A 5000 lb lot of crushed No. 8 road aggregate was produced with the bench-scale unit using FGD sludge from the AEP Gavin Station. Properties of the aggregates are compared with a commercial No. 8 gravel in Table 6. Also shown for comparison are AASHTO M-283 specifications for No. 8 size Class A aggregate. The gravel, provided by Trumbull Corp., was used in the road paving demonstration. Trumbull Corp. assisted in asphalt concrete design and road construction. Trumbull completed the Marshall stability test on the mix design with the 50/50 blend of manufactured aggregate and gravel (dry basis) and on the straight gravel as the coarse aggregate component in the asphalt concrete. The blend gave a higher Marshall stability (2400 lb vs. 2175 lb), higher air voids (4.4% vs. 3.4%), lower specific gravity (2.085 vs. 2.305), and lower flow (10.8 vs. 11.0) than the conventional material. The optimum asphalt dosage is 7.5 wt % with the blend and 6.8 wt % with the conventional aggregate; however, because the blend has a 11% lower specific gravity than the conventional material, the asphalt dosage on a volume basis is the same for the blend and the commercial mix. On October 20, 1998, a test pavement with CONSOL's manufactured aggregate was laid in the city of Warren, Trumbull County, OH. The manufactured aggregate was incorporated into the test asphalt concrete in a hot-mix asphalt plant operated by Lindy (a subsidiary of Trumbull Corp.) in East Brook, PA, for the pavement demonstration. Because of the higher moisture content of the as-produced manufactured aggregates relative to conventional aggregate, Lindy pre-dried the manufactured material before conducting the normal asphalt mixing operations. Lindy personnel reported that, other than the pre-drying step, the manufactured aggregate behaved similarly to conventional materials in that operation.

The test asphalt concrete is composed of 22.3% coarse manufactured aggregate, 22.3% coarse crushed gravel, 48.4% fine aggregate and 7.0% PG-55 (AC-20) asphalt. The manufactured aggregate constituted 50 wt % of the coarse aggregate in the mix (as-is basis); the asphalt, fine aggregate and the remaining 50% of the coarse aggregate were conventional materials. The conventional coarse aggregate was crushed gravel meeting PennDOT Type A No. 8 specifications and having a skid resistance level of excellent. Lindy milled and prepared the edges of a section (72" long x 10.8' wide x 1.5" deep) of the west-bound lane of North River Road NW in front of the Trumbull County Engineers' Office in Warren, OH. This section of highway is straight and nearly level. The average daily traffic count is about 11,000 vehicles per day. All indications are that the road bed is in excellent condition. The test asphalt concrete was used as a surface course to pave the prepared area. Conventional paving and rolling equipment was used. The edges were sealed, then the lane was opened to traffic. As a control test, a similar asphalt concrete, but made entirely with the conventional materials, was used to pave a similarly

prepared and sized area of the same lane. About 11.5 ton of the conventional asphalt concrete was required to pave the control test area. In contrast, only about 9.5 tons of the test asphalt concrete was required for the same sized area with manufactured aggregate; this reflects the low density of the manufactured aggregate. In other respects, the test concrete behaved similarly to conventional asphalt concrete during the paving operation.

Since November 1998, the structural integrity of test pavements and aggregates have been monitored. No aggregate degradation was identified until the second half of January 1999. The aggregate in the manufactured aggregate test pavement experienced some delamination, which could have been caused by the severe winter weathering and by the anti-skid materials (salt and slag) spread on the road for de-icing. However, the structural integrity of the test pavement remained good. No rutting and cracking was observed. Core samples taken from manufactured and natural aggregate test pavements in early February showed that the manufactured aggregate had an attrition rate twice that of the natural aggregate. The attrition rate was determined by filling the void volume in the surface of the core samples with ASTM -200 mesh standard sand (specific gravity of 3.188) and by measuring the volume of the sand. Core sample density measurements showed that the manufactured aggregate test pavement achieved only 90% compaction during construction, whereas the natural aggregate pavement met the target of 95% compaction. The higher attrition rate of the manufactured aggregate could be due to aggregate properties and insufficient pavement compaction during construction.

Since February 1999, no further aggregate degradation has occurred. The performance of the test pavement and the aggregate will be monitored for an additional year.

Concrete Block Manufacture. A 4000 lb batch of lightweight aggregate was produced from Cinergy Zimmer Station FGD sludge and used to manufacture concrete block. Properties of the crushed manufactured aggregates are compared with ASTM C331 lightweight aggregate specifications in Table 7. As shown in the table, the aggregate properties met the ASTM C331 aggregate specifications.

On November 23, 1998, concrete blocks were made with FGD sludge lightweight aggregate at the Walden Industries block plant in Tiltonville, OH. A total of 284 concrete blocks was produced in two batch runs (Batch 1 and 2) with a mix of 44.3 wt % manufacture aggregate, 43.8 wt % concrete sand and 11.9 wt % type 1 portland cement, and associated water. Batch 2 had a slightly higher water addition than Batch 1. The blocks have two hollow cores, they are 53.8% solid, and they have nominal dimensions of 8" x 8" x 16". Made for comparison were lightweight concrete blocks (made with a mix of Haydite (a commercial lightweight aggregate), grits, and cement), and normal weight blocks (made with a mix of sand, grits, and cement). All of the production runs were successful. The properties of the blocks are shown in Table 8. As indicated in Table 8, FGD sludge aggregate blocks from batches 1 and 2 met the ASTM C-90 lightweight concrete block specifications for water absorption (16.3% and 16.1% vs. 18%) and compressive strength (2117 psi and 2370 psi vs. 1900 psi), but slightly exceeded the maximum dry unit weight (106.7 lb and 106.9 lb/ft<sup>3</sup> vs. 105 lb/ft<sup>3</sup>). An increase in the relative amount of lightweight aggregate in the mix would result in the production of blocks meeting the lightweight specification. The FGD sludge manufactured aggregate block had a higher strength than commercial lightweight block and a lower strength than normal weight block. The compressive strength was determined with the use of end caps in accordance with the ASTM C90 test procedure.

On December 14, 1998, Walden Industries constructed a fence wall in Tiltonville, OH, with FGD sludge aggregate block, Haydite lightweight aggregate block, normal weight aggregate block, and Belite (bottom ash) block to determine the performance of the aggregate in actual use. Each block section has a dimension of 6' x 25'. Belite block is a commercial medium weight block sold by Walden Industries for use in construction.

Since December 1998, the performance of the fence wall made with manufactured lightweight aggregate block has been monitored. No structural degradation was observed. The section of test wall made with CONSOL's manufactured aggregate blocks performed as well as wall sections made with commercial normal weight aggregate and Belite blocks. The performance of fence wall and manufactured lightweight aggregate will be monitored until December 2000.

### Pilot Plant Demonstration

U. S. DOE, Duquesne Light Co., SynAggs, Inc., and CONSOL R&D co-funded the construction and operation of a 500 lb/hr integrated, continuous pilot plant in 1999. The pilot plant was designed as a scale-up of the bench-scale unit. Lime wet FGD sludge from Duquesne Light Elrama Station was used for aggregate production in the pilot plant. The pilot plant demonstration was completed on July 1, 1999, after six weeks of operation. About 135 tons of aggregate products, including lightweight aggregate, medium weight aggregate, and road aggregate, were produced with different mix formulations. The pilot plant test successfully demonstrated the continuous, integrated operation of the mixing, pelletizing and curing steps, and demonstrated process flexibility to produce various products. In addition, the pilot plant provided operating data and experience sufficient to design and cost a commercial plant. The pilot plant demonstration was a major step toward commercialization of manufactured aggregate production from FGD materials.

It is planned to use the pilot plant to produce large quantities of aggregates from other sources for product demonstration and further process development.

### **ACKNOWLEDGMENT**

This work was supported by the Ohio Coal Development Office (Grant Agreement Nos. CDO/D-902-9 and CDO/D-95-2), the U. S. Department of Energy (Cooperative Agreement No. DE-FC26-98FT40027), Trumbull Corp., SynAggs, Inc., Duquesne Light Co., Garick Corp., Walden Industries, Inc., and CONSOL Inc.

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**TABLE 1**

**COMPARISON OF FBC ASH AGGREGATE PROPERTIES WITH AASHTO CLASS A AGGREGATE SPECIFICATIONS**

	FBC Ash Aggregates	Class A Aggregates AASHTO M-283 No. 8 Size
LA abrasion index, wt %	19	40 (max)
Soundness index, wt %	11	12 (max)
Unit weight, lb/ft <sup>3</sup> , as det.	75	---
Unit weight, lb/ft <sup>3</sup> , dry basis	65	70 (min) <sup>(a)</sup>
fractured pieces, wt %	70	varies, depends on State regulation
<u>grain size, wt % passing</u>		
2"	100	100
3/8"	89.5	85 - 100
4 mesh	12.9	10 - 30
8 mesh	0	0 - 10

(a) Unit weight specification applies only to slag in AASHTO M-283

**TABLE 2**

**MARSHALL STABILITY TEST RESULTS FOR ASPHALT MIXES  
MADE WITH FBC ASH AGGREGATES**

Mix	A	B	C	D	E
<u>Mix Composition</u>					
FBC Ash Aggregates, wt %	7.9	16.7	38.5	38.0	37.5
Mfd. Limestone Sand, wt %	62.5	60.6	56.0	56.0	56.0
No.8 Limestone, wt %	23.6	16.7	0.0	0.0	0.0
Asphalt Cement, wt %	6.0	6.0	5.5	6.0	6.5
<u>Test Results</u>					
Marshall Stability, lb	2250	2325	2875	2305	2275
Flow	11	12	14.5	15	15.3
Bulk Specific Gravity	2.236	2.154	1.934	1.936	1.927
Air Voids, %	7.4	10.7	16.1	15.5	15.4
Voids Mineral Aggregate, %	17.4	19.6	21.8	22.1	22.8

**TABLE 3**

**SIZE DISTRIBUTION OF THE THF-EXTRACTED CORE SAMPLES  
AND ESTIMATED SOURCE OF FINES**

	1995							1996
	6/1	6/27	8/3	9/1	10/2	11/1	12/4	5/30
<b><u>Size Dist., wt %</u></b>								
+3/8"cm	4	13	9	11	9	6	12	7
3/8" x 4 mesh	45	36	43	42	42	44	31	36
4 x 8 mesh	17	16	15	13	15	16	17	21
8 x 50 mesh	18	19	15	13	13	14	17	20
-50 mesh	16	17	18	21	21	20	23	16
<b><u>Source of fines, wt %</u></b>								
<u>8 x 50 mesh</u>								
FBC ash aggregates	-	35	33	30	36	31	31	31
CaCO <sub>3</sub>	-	48	50	53	50	51	51	49
Extraneous	-	17	17	17	14	18	18	20
<u>-50 mesh</u>								
FBC ash aggregates	-	33	31	29	33	35	29	27
CaCO <sub>3</sub>	-	41	40	42	42	38	42	42
Extraneous	-	26	29	29	25	27	29	31

**TABLE 4**

**PROPERTIES OF LIGHTWEIGHT AGGREGATES  
PRODUCED FROM SPRAY DRYER ASH**

	<b>Spray Dryer Ash Aggregates, First batch</b>	<b>Spray Dryer Ash Aggregates, Second Batch</b>	<b>Lightweight Aggregate Specifications ASTM C-331</b>
Grain Size	Meets ASTM No. 8 -	- Meets ASTM combined Nos. 8/9	No. 8 (coarse) Nos. 8/9 (combined)
Dry Unit Weight, lb/ft <sup>3</sup>	52.6 (No. 8) -	- 55.4 (Nos. 8/9)	No. 8 = 55 (max) Nos. 8/9 = 65 (max)
Clay Lumps, wt %	1.7	1.5	2.0
Staining Material	negative	negative	negative

**TABLE 5**

**PROPERTIES OF CONCRETE BLOCK MADE FROM SPRAY DRYER ASH AGGREGATE**

	<b>Batch 1 Blocks</b>	<b>Batch 2 Blocks</b>	<b>ASTM C-90 Lightweight Concrete Block Spec.</b>
Unit Weight, lb/ft <sup>3</sup> , dry	99.9	100.9	105 (max)
Water Absorption, wt %	16.9	16.8	18 (max)
Net Compressive Strength, psi	2953	1930	1900 (min)
Deformation at Failure, %	1.9	1.9	-

**TABLE 6**

**COMPARISON OF FGD SLUDGE AGGREGATE PROPERTIES WITH GRAVEL AND  
AASHTO CLASS A AGGREGATE SPECIFICATIONS**

<b>Properties</b>	<b>Manufactured Aggregate</b>	<b>Gravel</b>	<b>Class A No. 8 Aggregates</b>
LA Abrasion Index, wt %	26	27	40 (max)
Soundness Index, wt %	0 <sup>(a)</sup>	4.1	12 (max)
Unit Weight, lb/ft <sup>3</sup> , as det	71	99	---
Unit Weight, lb/ft <sup>3</sup> , dry basis	62	98	70 (min) <sup>(b)</sup>
<u>Grain Size, wt % passing</u>			
2"	100	100	100
3/8"	100	86	85-100
4 mesh	23	22	10-30
8 mesh	60	7.9	0-10

- (a) Determined in accordance with ASTM C-88 procedure. The index value is sensitive to sample holder characteristics (size, shape and construction material) that are not specified in the method. Therefore, the value can range widely for a given material.
- (b) Unit weight specification applies only to slag in AASHTO M-283.

**TABLE 7**

**PROPERTIES OF LIGHTWEIGHT AGGREGATE MADE FROM CINERGY ZIMMER STATION FGD SLUDGE**

<b>Property</b>	<b>FGD Sludge Aggregate</b>	<b>Lightweight Aggregate Specifications ASTM C-331</b>
<u>Unit Weight, lb/ft<sup>3</sup></u>		
As Is	60	---
Dry	50	55 (Coarse, Max) 65 (Combined, Max)
<u>Clay Lumps, wt %</u>	1.7	2.0
<u>Staining Material</u>	Negative	Negative
<u>Particle Size, wt % pass</u>		
3/8"	100	Nos. 8/9 (Combined)
4 mesh	72.8	
8 mesh	40.7	
16 mesh	27.0	

**TABLE 8**

**PROPERTIES OF FGD SLUDGE, NORMAL WEIGHT AND HAYDITE LIGHTWEIGHT BLOCKS**

	FGD Sludge Blocks		Normal Weight Block	Haydite Lightweight Block	ASTM C-90 Lightweight Block Spec.
	Batch 1	Batch 2			
Unit Weight, lb/ft <sup>3</sup> , dry	106.7	106.9	126.9	98.3	105 (max)
Water Absorption, wt %	16.3	16.1	9.8	14.1	18 (max)
Net Compressive Strength, psi	2117	2370	2727	1750	1900 ( min)