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Particulate Hot Gas Stream Cleanup Technical Issues

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Particulate Hot Gas Stream Cleanup Technical Issues

CONTRACT INFORMATION

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Period of Performance	October 1994 to September 1996

The nature of the collected ash has been identified as an issue creating barriers to the commercialization of advanced particle control technologies. Since most of the emphasis and extended operation of Hot Gas Stream Cleanup (HGCU) facilities have been with ceramic candle filters, problems with ash characteristics can be understood in terms of their effects on these control devices. This project is designed to identify the ways ash characteristics affect advanced particle control technologies, to construct and maintain a data base of HGCU ashes and their measured characteristics, and to relate these characteristics to the operation and performance of these facilities.

The key characteristics of the collected ash are the morphology of the overall ash aggregate (porosity, geometry of the pores, specific surface area, etc.), and the cohesivity of the aggregate. Cohesivity is controlled in turn by the morphology of individual particles (size, shape), and the strength of the forces between particles due to sintering, chemical bonds, van der Waals forces, adsorbed liquid layers, and salt bridges.

Our data base currently comprises 242 ash samples from 12 combustion and gasification HGCU sources. We have 116 ash samples from the Advanced Particulate Filter (APF) at the Pressurized Fluidized-Bed Combustor (PFBC) at Tidd, 51 ash samples from the Pressurized Circulating Fluid Bed (PCFB) system located at Karhula, Finland, and 17 ash samples from Foster Wheeler's Second Generation PFBC system at Livingston, New Jersey. In addition to these three operating facilities, our stock of HGCU samples includes ashes from facilities operated by Grimethorpe, Westinghouse, MW Kellogg, New York University, the University of North Dakota Energy and Environmental Research Center (UNDEERC), and KRW. We have characterized aerodynamic size distribution, specific surface area, uncompacted bulk porosity, tensile strength, true particle density, drag-equivalent diameter, specific gas flow resistance, nodule porosity, particle morphology (with Scanning Electron Micrographs), and mineral composition of selected ashes from advanced particle control devices. Ranges of values of several of these key quantities are shown in Table 1.

Table 1
Ranges of Characteristics of HGCU Samples

quantity	HGCU ashes
Stokes' MMD, μm	0.74 - 60
filter cake porosity, %	57 - 84
specific surface area, m^2/g	0.5 - 353
uncompacted bulk porosity, %	62 - 97
specific gas-flow resistance at uncompacted bulk porosity, in $\text{H}_2\text{O}/(\text{ft}/\text{min})/(\text{lb}/\text{ft}^2)$	0.8 - 61
drag-equivalent diameter, μm	0.08 - 9.0

In general, the gasification samples we have analyzed have very high specific surface areas. Because filtering drag is accumulated as the gas being filtered passes over the surfaces of the particles in the filter cake, high specific surface areas generally correlate with small values of drag-equivalent diameter. (Drag-equivalent diameter incorporates the effects of particle morphology on filtering drag.) The effect of filter cake structure on filtering drag is determined by the cake porosity. Therefore, filtering drag is a function of the shape of the particles in the filter cake and the porosity of the cake.

Experiences at the Tidd APF

Observations of the Advanced Particulate Filter (APF) at the Tidd PFBC Demonstration Plant led to the conclusion that tenacious ash deposits had formed in the filter vessel and induced stresses that resulted in bent or broken ceramic candle filter elements. The proximity of these bent and broken candle filter elements to large, strong ash deposits emphasized the need to prevent or control the growth of these deposits, facilitate their on-line removal, and/or to develop filter design criteria to minimize their effects on individual filter elements. Similar,

although less severe, problems have also been observed at the PCFB facility located in Karhula, Finland.

Tidd ash chemistry combined with the environment within the filter vessel caused ash deposits formed in the filter vessel to consolidate and strengthen. We believe the strengthening of these deposits is due to formation of eutectic compounds. These eutectics form when primary coal ash particles (containing a large percentage of aluminosilicate compounds) come into physical contact with sorbent-derived ash particles which contain relatively large amounts of magnesium and/or calcium. Aluminosilicate compounds in the coal ash particles react with alkali metals in the sorbent ash particles to form eutectics that melt at relatively low temperatures. This reduction in melting points combined with long-term exposure to the temperatures in the filter vessel tends to create relatively soft, sticky layers on the surfaces of the ash particles. The surface tension of the near-liquid layer on the particles pulls adjacent ash particles closer together, thereby eventually consolidating the structure of the entire ash agglomerate. The progress of these reactions is supported by the intimate contact of the ash particles in the agglomerate and by long-term exposure of the ash to the temperatures in the filter vessel.

The optimum solution to the problems caused by these ash aggregates is the removal of these aggregates from the filter before the eutectics have had enough time to develop. The approach that has proved most successful at Tidd for eliminating the deposits was the total bypassing of the cyclone upstream of the APF. This increased the size distribution of the particles forming the various ash deposits, thereby decreasing their inherent cohesivity. These agglomerates of lower cohesivity did not have sufficient strength to remain in the APF long enough to undergo consolidation.

Recent Analyses of HGCU Ashes

Table 2 lists three samples we have recently analyzed and that we discuss in this paper.

Table 2
Selected Samples from the HGCU Data Base

ID #	Source	Brief description
4170	DOE/METC	pilot-scale gasifier
4176	UNDEERC	TRDU P047: 4/18-20/96
4182	Karhula	middle plenum ash 1996

The first sample listed in Table 2 was obtained from the Modular Gas Cleanup Rig (MGCR) gasification facility located at DOE/METC. This sample was collected from the hopper of the pilot-scale candle filter assembly. The second sample listed in Table 2 was received from the Transport Reactor Demonstration Unit (TRDU) located at UNDEERC. This sample was identified as TRDU P047 collected from 4/18/96 to 4/20/9. Information provided by UNDEERC indicated that the TRDU operated for a total of 117 hours under gasification conditions in April. The TRDU conditions during these tests were described as not sustainable for extended use. We proceeded with a full analysis of this sample despite the limitations of the conditions under which it was produced. We also received six ash samples from the PCFB facility located in Karhula, Finland. The samples included filter cake ashes collected from the top, middle, and bottom plenum candle surfaces, as well as hopper and bottom ash samples. We selected the sample of filter cake ash from the middle plenum for detailed analysis. The results of our chemical and physical analyses of these samples are summarized in Tables 3 and 4 and discussed in the following sections.

Table 3
Chemical Analyses of Selected Samples, % wt.

constituent	ID #	4170	4176	4182
Li ₂ O		0.02	0.01	0.01
Na ₂ O		0.59	0.63	0.87
K ₂ O		0.07	0.12	1.6
MgO		10.9	5.2	0.72
CaO		33.3	11.2	17.4
Fe ₂ O ₃		1.17	1.9	11.0
Al ₂ O ₃		17.4	22.9	12.6
SiO ₂		31.8	54.8	34.4
TiO ₂		1.49	0.8	0.6
P ₂ O ₅		0.53	0.55	0.10
SO ₃		0.32	0.96	19.8
LOI		35.9	4.3	0.22
soluble SO ₄ ⁼		<0.2	--	23.8
Equilibrium pH*		10.2	--	7.4

* Equilibrium pH is dimensionless.

Table 4
Physical Characteristics of Selected Samples

quantity	ID #	4170	4176	4182
Stokes' MMD, μm		0.74	60	9.9
filter cake porosity, %		--	--	75
specific surface area, m^2/g		140	51	1.2
uncompacted bulk porosity, %		97	72	81
specific gas flow resistance, at uncompacted bulk porosity, in $\text{H}_2\text{O}/(\text{ft}/\text{min})/(\text{lb}/\text{ft}^2)$		18	2.8	1.5
drag-equivalent diameter, μm		0.08	2.77	2.22
tensile strength, N/m^2		0.6	2.8	3.8
true particle density, g/cm^3		2.87	2.60	2.83

DOE/METC MGCR

Of all the gasification samples in our HGCU data base, the sample exhibiting the lowest permeability to gas flow (or the highest specific gas flow resistance) was the sample from the DOE/METC MGCR (ID # 4170). Even though this sample would be expected to initially form a

filter cake with a porosity on the order of 97 % (the uncompacted bulk porosity value measured for this sample), the morphology of the particles in this sample, specifically their small size, is the ultimate cause of its high resistance to filtering flow. Although other samples in the HGCU data base have higher values of specific surface area than this sample, it exhibited the lowest values of physical particle size and drag equivalent diameter of all the gasification samples we have tested. When the DOE/METC MGCR sample was examined with a scanning electron microscope (Figure 1), the fineness of its particle size distribution was readily apparent. Like other gasification samples, another distinctive characteristic of the DOE/METC MGCR sample was its high value (35.9 % by wt.) of loss-on-ignition (LOI).

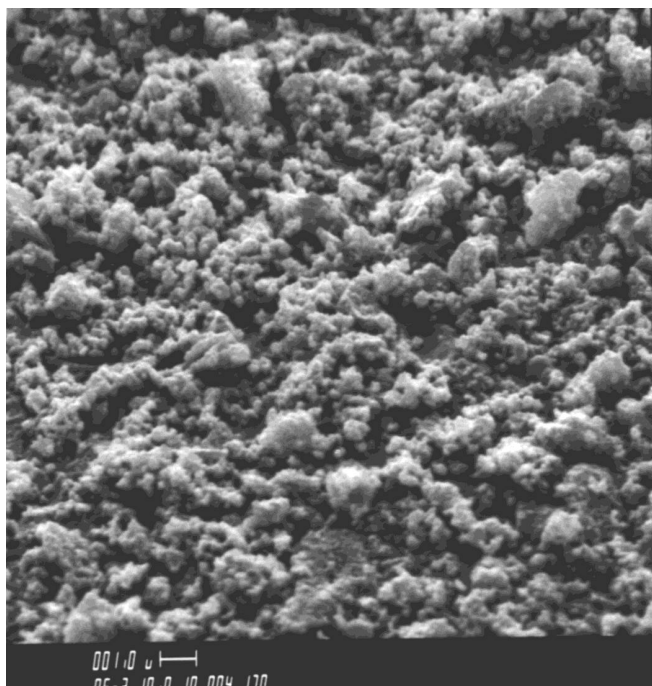


Figure 1. SEM photograph of DOE/METC MGCR gasification residue.

We also characterized the response of the DOE/METC MGCR sample to compacting forces. The compaction data, which we measured at room temperature, show that a filtering pressure drop of 3.4 psi (94 in. H₂O)

may be sufficient to reduce the porosity of a filter cake formed from this sample down to about 90 %. Data from our permeability model indicate that the degree of filter cake consolidation that may be induced by normal filtering pressure drops of around 3 to 4 psi may increase filtering pressure losses by a factor of twenty or more. This is especially detrimental since the specific gas-flow resistance of this sample is quite high even when the filter cake is uncompacted. (The specific flow resistance of the MGCR sample is 18 in H₂O/(ft/min)/(lb/ft²) for a filter cake having a porosity of 97 %. If a filter cake composed of this ash is compacted to around 90 % porosity, specific gas flow resistances around 400 in H₂O/(ft/min)/(lb/ft²) could be expected.) These characteristics ultimately lead to high pressure losses and/or very frequent cleaning. Although the METC MGCR sample showed an apparent increase in strength as it was compacted, the cake compacted with a pressure of 3.4 psi still had comparatively low strength.

Overall, our observations of the DOE/METC MGCR sample and other gasification samples indicate that the filtration of gasification particulate residues can generate filter cakes that exhibit extremely low permeabilities. These low permeabilities can be traced to the presence of a high proportion of submicron particles and/or extremely high specific surface areas. In addition, filter cake compaction may exacerbate these negative characteristics. Consequently, it may be difficult to maintain a reasonable pressure drop in their filtration.

In general, the gasification samples we have tested exhibited very high uncompacted bulk porosities, which indicates that they are highly cohesive. (High uncompacted bulk porosities are generally associated with samples having fine size distributions and/or irregular particle shapes. Gasification and PFBC often generate both of these characteristics.) However, gasification samples also exhibit relatively low

tensile strengths. Normally, we would expect that highly cohesive powders would also have high tensile strengths. We are not yet certain what causes this anomaly with the gasification samples. The low tensile strengths we have measured for these samples may indicate that ash dislodged from filter elements during pulse cleaning cycles may break up into very small agglomerates. If this type of breakup occurs, reentrainment of previously collected gasification residues may pose a significant problem.

UNDEERC TRDU

The TRDU ash (ID # 4176) is very coarse, free flowing, has a high bulk density, and is medium gray. These observations agree with the quantitative evaluations we performed. SEM photographs of this ash have shown a large proportion of spherical particles, suggesting that temperatures above the fusion temperature of the ash were present in the TRDU. (We have not observed spherical particles in any of the other HGCU ashes in our data base.) This ash sample exhibited a relatively large drag-equivalent diameter and a low specific flow resistance, despite its relatively high specific surface area. These data suggest that most of the surface area is attributable to very fine pores. Because the gas flowing through a filter cake never enters extremely fine pores on the surfaces of the collected particles, the particle surface area contained in these pores does not contribute substantially to filtering pressure drop.

Karhula PCFB

Figure 2 presents an SEM photograph of a fresh fracture surface (ID # 4182) of a filter cake nodule taken from the filters at Karhula. This photograph indicates that the nodules found in the Karhula filter are concretions composed of discrete fine particles almost completely embedded in pervasive amorphous masses which apparently form in the filter vessel after

the particles are initially collected. The appearance of the Karhula filter cake nodule in Figure 2 is very similar to the appearance of nodules removed from the Tidd APF. This similarity, in combination with similar ash chemistries and flue gas environments, lead us to believe that eutectic formation, as described above in the section summarizing our observations from the Tidd APF, is also responsible for nodule formation at Karhula.

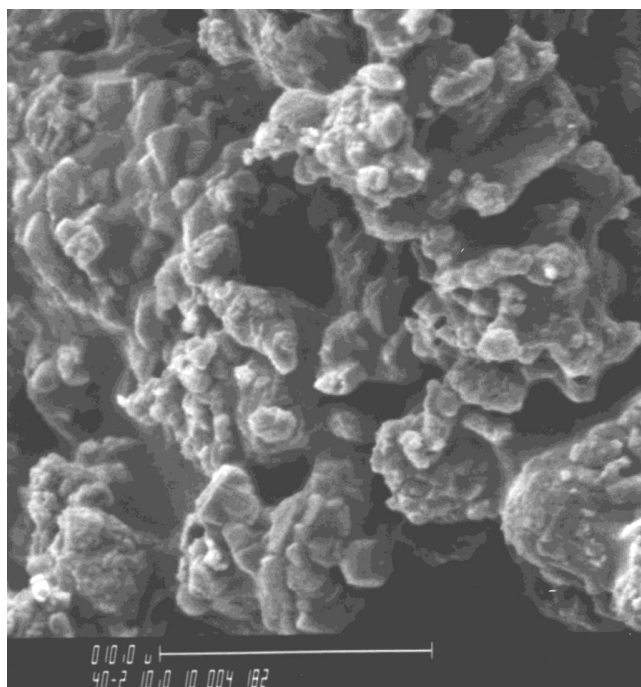


Figure 2. SEM photograph of a fresh fracture surface of a Karhula filter cake nodule.

The filter cake ash samples we received from Karhula each contained fragments of nodular filter cake. We used one of the methods we developed for measuring the overall pore volume of a nodule to characterize the nodule fragments in sample # 4182. Our on-site experience at the Tidd PFBC has shown that the porosity of the most recently deposited portions of filter cake is significantly greater than the porosity of older portions of the cake. It is very likely that during the shipment of the filter cake ash and nodule samples from Karhula to our laboratories, the fluffiest, most recently

deposited parts of the cake were shaken or rubbed off of the cake nodules. Therefore, the reported filter cake porosity value of 75 % represents a lower bound for the actual overall filter cake porosity.

HGCU Data Base Development

We have designed the primary structure of the HGCU data base that we are constructing in Microsoft Access™. Access will allow the user to compose various graphs and data presentations based on filtered and sorted groups of data. We have scanned in SEM photographs of the samples in the data base and still images taken from the videotapes we made during our four site visits to the Tidd PFBC. In addition to measured characteristics of the samples, we intend to include background information related to the various facilities such as the participating organizations, the key operating personnel, process descriptions, photographs of the facility, and literature citations. We are designing a variety of summary reports that the data base user will be able to view and print. We plan to issue the HGCU data base as a run-time version of Microsoft Access stored in CD-ROM format. This format will be required since the final data base will contain a large number of photographic images that have significant storage requirements.

High Temperature Ash Analyses

To properly assess the role of temperature and flue gas constituents on the key characteristics of the collected ash, these characteristics should be determined for samples that have been conditioned and tested in environments as much like those found in the filter vessel as possible. Therefore we plan to design, construct, and evaluate two devices that will allow measurements of the tensile strength, permeability, and uncompacted bulk porosity of samples at temperatures up to 1650 °F and in simulated flue gas environments. Our plans do

not include performing these tests at elevated pressures.

The tensiometer we currently use to characterize samples at temperatures up to 300 °F operates on the principle of inducing a charge on the surface of an sample layer prepared on the grounded electrode of a parallel-plate high voltage arrangement¹. As the electric field between the plates is increased, the attraction between the charges induced on the particles on the surface of the sample and the electrical potential of the opposite electrode eventually exceeds the force holding the charged particles to the rest of the sample. At this point the particles are ejected from the sample on the grounded electrode and travel to the opposite electrode. The electrical force required to separate these particles from the rest of the sample is equal to the tensile strength of the sample as it exists on the grounded plate. We plan to select appropriate materials and modify our design as required to construct a tensiometer that can operate at temperatures up to 1650 °F and that will be capable of conditioning and testing powder samples in simulated oxidizing or reducing flue gas environments.

We also plan to build a device to measure uncompacted bulk porosity and sample permeability at temperatures and in gases with compositions characteristic of HGCU devices. This device will consist of a vertical cylinder (approximately 20 cm in diameter) with a transparent quartz top and a porous disk (possibly a ceramic substrate like those used in the patch tester) near the bottom through which gas can flow. A small plenum on the other side of the porous disk will be connected to a heat exchanger and downstream flow measuring equipment. The device will be fitted with ceramic heaters that will maintain the device at temperatures up to 1650 °F.

Permeability measurements will be made as a function of the thickness of the sample in the isolation ring. We will measure the pressure

drop across the sample at various gas flow rates. The filtering pressure drop across the sample at a given flow rate will be used to compact the sample prior to measurement of sample permeability. Repetition of this measurement cycle will result in a series of data values relating filtering drag to sample porosity. Thus, with this device it will be possible to compare these values determined at ambient conditions with values measured at temperatures and with gas compositions typical of HGCU operation.

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Reference

1. Pontius, D.H. and T.R. Snyder, "Measurement of the tensile strength of uncompacted dust aggregates," Powder Technology, 68 (1991) pp. 159-162.