Development of a Monolithic Ceramic Cross-Flow Filter: Part 2

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Introduction

Ceramic cross-flow filters were designed to provide high temperature, high pressure removal of particulates from hot gas streams in coal fueled power systems, with the benefit of high surface area per unit volume. An important need has been identified as a one-piece monolithic ceramic cross-flow filter, rather than the most recent segmented filter that has been known to delaminate/separate in service. Current ceramic technology has proved incapable of forming a complex one-piece shape such as this, with an acceptable permeable, porous ceramic material. Blasch's unique and proprietary injection mold ceramic forming process has commercially produced complex shapes of this nature for many years, but of a non-permeable ceramic. In the earlier SBIR Phase I work, permeable, porous ceramic compositions were developed, targeted to meet the filtration and other requirements of cross-flow filters. In this SBIR Phase II effort, the ceramic composition data and lab scale process techniques established in Phase I are being used as the basis for prototype production of full-size monolithic ceramic cross-flow filters. After production, these prototypes will be subjected to filtration testing, followed by post-test characterization.

Progress thus far in Phase II (through the first half of the project) includes design of a prototype full size monolithic ceramic cross-flow filter, acceptable material screening tests including room temperature permeability and particulate collection efficiency, design and fabrication of a mold, development of a larger scale process to produce prototypes, and production of some initial prototype filters.

Objectives

The overall primary objective of this Phase II project is to use the ceramic composition data and lab scale process techniques established in Phase I for prototype full scale production and filtration testing/ characterization of monolithic ceramic cross-flow filters. The specific project objectives for the duration of this project, simply stated, are:

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1. Design a prototype full size one-piece monolithic cross-flow filter ceramic shape to be produced using the Blasch process and permeable ceramic compositions developed in Phase I.

2. Design and fabricate a mold system capable of forming this prototype full size monolithic ceramic cross-flow filter.

3. Characterize permeable ceramic compositions to be used for this prototype filter, to include room temperature permeability, particulate collection efficiency, and corrosion testing.

4. Produce prototype full size monolithic ceramic cross-flow filters by upscaling the lab scale process developed in Phase I.

5. Evaluate performance of these prototype filters in filtration testing and characterize after testing.

Approach

The detailed design of a prototype full size monolithic ceramic cross-flow filter was first determined before a mold system could be designed for a complex shape of this nature. This design had to include consideration of fulfilling the application requirements, as well as manufacturability. Knowledgeable people from Westinghouse and DOE-METC provided input to Blasch regarding design criteria for this prototype filter. Dimensional tolerances for the filter part were also determined as part of this design. Various clean-gas and dirty-gas channel layouts and sizes were considered, to yield a monolithic cross-flow filter that has beneficial filtration surface area, is manufacturable, and is not likely to plug during hot gas filtration application.

This monolithic cross-flow filter design was used as the basis for the design of the mold system required to form this filter prototype shape. This mold system was designed to be useable in Blasch's process and to yield the tight dimensional tolerance requirements of the filter design. Once this mold system was designed, fabrication was performed.

A fairly simple mold was also designed and fabricated to form the different sized ceramic disk specimens needed for room temperature permeability testing, particulate collection efficiency testing, and corrosion testing, all by Westinghouse. These specimens were made using the technique and composition(s) developed during Phase I..

It was realized that the production of prototype monolithic ceramic cross-flow filters would require some equipment of a larger size than the lab scale equipment used in Phase I. Blasch already possessed most of the necessary equipment, but needed a larger mixing apparatus for hi-speed mixing of the constituents of the permeable compositions developed in Phase I, including organic additives.

Engineering was performed to determine appropriate type, size, and speed of mixer needed for this process step. Subsequently, this mixing apparatus was procured. After installation of this equipment, mixing parameters were determined (such as time, speed, and batch size) by trials, and microscopic comparison of the same slurry composition mixed on a much smaller, lab scale.

Ceramic raw materials were and are being prepared, including sieving, to obtain particles of the proper size. Kiln furniture setter plates, on which to set the ceramic cross-flow filters during firing were specified and procured. These plates must handle the specified firing conditions of the filters, and also be very flat and resist hot load deformation to assure that the prototype filters are made with flat surfaces.

The lab scale process used in Phase I to make small test specimens was upscaled in this Phase II effort to produce monolithic ceramic cross-flow filters. Specific batching, mixing, and forming method details were planned based upon past upscaling experience, including the Phase I work. This upscaled method was and is subject to modification based upon the experience gained while trying to produce these ceramic cross-flow filters. Drying and firing cycles were also determined, to assure that the filter prototypes do not deform, crack, or explode during drying or firing. Several prototype filters are likely to be produced while streamlining the process, in attempt to make filters that meet the permeability, surface quality, dimensional, and other requirements.

Finally, three prototype full sized monolithic cross-flow filters each of two different permeable ceramic compositions will be submitted to Westinghouse. Filtration testing of two filters each of two compositions (the remaining one filter of each composition will be retained by Westinghouse as a reference for later comparison) will be performed by Westinghouse, followed by full characterization of the filters **after** this testing (post-test characterization). The filtration testing will occur in Westinghouse's HTHP PFBC simulated test vessel, at "typical conditions". These conditions include the following: 845þC temperature, 10 atm pressure, combusted natural gas atmosphere, with gas velocity of 10 fpm. The filters will be operated for a period when ash is delivered to form the dust cake along the filter surface. The cake will then be removed by pulse cleaning. Various measurements will be taken during the test.

Project Description

Blasch's unique and proprietary injection mold ceramic forming process has commercially produced complex shapes for many years, but of a non-permeable nature. In the earlier SBIR Phase I work, permeable, porous ceramic compositions were developed, targeted to meet the filtration and other requirements of cross-flow filters. In this Phase II effort, the Blasch forming process is being used as the basis along with the ceramic composition data and lab scale process techniques established in Phase I, for prototype production of full size monolithic ceramic cross-flow filters.

Generally speaking, the proprietary Blasch forming process includes the following steps: batching/weighing, wet batch mixing, injection of ceramic slurry into molds, solidification by freezing, removal of mold, drying, and firing. This same general process is being used to make these prototype monolithic ceramic cross-flow filters, but with modification based upon Phase I experience. This modification includes a more intensive mixing procedure, to assure the organic additives are well dispersed and mixed. Also, drying and firing cycles are different to accomodate the slow burn-out of these organics; otherwise the filter prototype could crack, deform, or even explode.

Permeable ceramic material compositions developed in Phase I are being used for these prototype filters. The alumina-mullite permeable ceramic compositions identified as #4-270 and #4-300 were selected based upon these Phase I results. These two are actually the same raw material composition, but are fired at different temperatures, resulting in different mullite contents. Composition #4-300 is the higher fired body, with the higher % mullite.

In accordance with the earlier stated "Approach", the following has been performed during the first half of this project:

1. A prototype full size monolithic cross-flow filter was designed that seems to be manufacturable. This design also seems to fulfill the stated criteria for the application with regards to high surface area of filtering walls (about 8.4 square feet), and large enough dirty gas channels to reduce plugging (0.30 inches high x 0.9 inches wide, and 0.30 inches high x 2.0 inches wide). Figure 1 in the results depicts a three-dimensional cut-away view of this monolithic cross-flow filter design.

2. A mold system for the filter prototype was designed and fabricated. The mold system was made with smooth surfaces and tight dimensional tolerances to accomodate the dimensional and surface finish requirements incorporated into the filter design.

3. A mold was designed and fabricated for the different sized ceramic disk specimens required for Westinghouse's room temperature permeability testing, particulate collection efficiency testing, and corrosion testing. Ceramic disk specimens of compositions #4-270 and #4-300 were formed, dried, fired and sent to Westinghouse for testing.

4. Westinghouse performed room temperature permeability and particulate collection efficiency testing on compositions #4-270 and #4-300. Results are discussed later in this paper. Corrosion testing on composition #4-300 is in process as this report is being written.

5. Mixing apparatus was investigated, engineering was performed, and a larger scale hi-speed mixer was procured. After installation, mixing parameters were determined, including speed and mixing time. This was done by microscopic comparison of permeable ceramic composition raw wet mix specimens mixed in this new mixer, compared to samples of the same composition mixed in the lab scale mixer used earlier in Phase I.

6. In effort to determine optimum processing techniques and parameters, several (ten at the time of this paper) prototype monolithic ceramic cross-flow filters were produced. All were dried and fired based on cycles that were determined earlier. Various ceramic forming technique

modifications were and are being tried to eliminate the voids on interior filter walls. These include use of different vibration techniques and composition moisture contents.

Results

Many of the results are obvious based upon earlier discussion of the project descripition. The prototype monolithic ceramic cross-flow filter was designed, and has approximate overall outside dimensions of 12 inches x 12 inches x 4 inches (not including the flange at the bottom). The intake "dirty gas" channels are 0.30 inches high x 2.0 (or 0.9) inches wide x 3.8 inches long. The exhaust "clean gas" channels run perpendicular to the dirty gas channels, and measure 0.14 inches high x 1.5 inches wide x 11.6 inches long. This design yielded filter surface area of about 8.4 square feet. Figure 1 is a three dimensional view of this filter design. Also, it is specified in this design that all internal filtering walls, which are nominally 0.11 inches thick, are to be free of voids larger than 0.025 inches diameter, such that the chance for a "blow-through" is minimized.

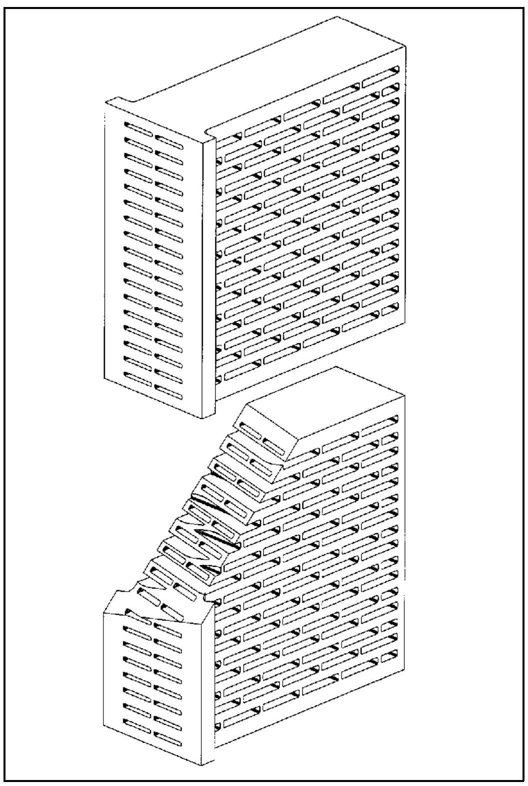

Figure 1. 3-D Depiction of Prototype Full Size Monolithic Ceramic Cross-Flow Filter

Westinghouse performed room temperature gas permeability and particulate collection efficicncy testing on disk specimens of compositions #4-270 and #4-300. Westinghouse indicated that the room temperature gas permeability results for both composition specimens was acceptable with regards to their target of 1 in-wg/fpm, as is shown in the following table.

Disk ID No.	Room Temperature Gas Flow Resistance, in-wg/fpm
BPC 4-270	1.06 - 1.13
BPC 4-300	0.93 - 1.01

In the particulate collection efficiency test, 75 grams of dust were fed to each of the two disk specimens (compositions #4-270 and #4-300). No observable dust was detected in the outlet gas during testing. Similarly, an approximate 1mm thick dust cake was formed along one surface of each disk, and when the disks were fractured, limited penetration of fines was detected in each matrix. From these qualitative observations, the dust cake layer formation and extent of penetration was considered to be acceptable by Westinghouse, in terms of particulate infiltration into the first pore layers of these porous ceramic bodies. Further, Westinghouse indicated that their gas flow resistance and particulate collection efficiency data imply that the Blasch oxide-based matrix was reproducibly manufactured for use in this effort. As this paper is being written, results from Westinghouse on corrosion resistance testing of disk specimen of composition #4-300 are not yet available.

Ten prototype full size monolithic ceramic cross-flow filters have been made thus far in this project. Some were made of composition #4-270 and others of #4-300. These met the prescribed dimensional tolerance criteria, had no sign of firing warpage or deformation, and had in general, had very good exterior appearance, but had minor voids (unacceptable) in the filtering walls. One filter did however have a deformed top area and another cracked badly: in both cases, the fault occurred because of identified specific variations in forming and mold-use techniques. Of the prototypes produced by Blasch thus far, the design criteria has been met as shown in the following table.

Filter No.	Compos-ition	Warp/ Deform	Meet Dimensional Tolerances?	Meet Void Specs?	Overall Appearance
4-A	4-270	none	yes	no	v. good
4-B	4-300	broke	yes	no	poor
4-C	4-270	none	yes	no	v. good
4-D	4-270	none	yes	no	v. good
4-E	4-300	none	yes	no	v. good
4-F	4-300	none	yes	no	v. good
4-GW	4-300-W*	some	yes	no	poor
4-HW	4-270-W*	none	yes	no	v. good
4-IW	4-270-W*	none	yes	no	v. good
4-JW	4-300-W*	none	yes	no	v. good

* "-W" denotes additional moisture added to wet mix.

Figure 2 is a photograph of one of the prototype monolithic ceramic cross-flow filters made in this project using the Blasch process. This filter is typical of those produced so far.

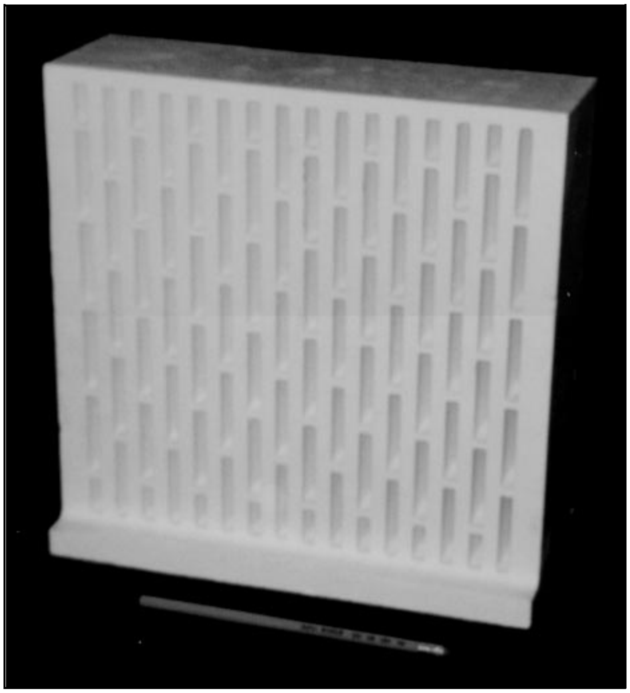

Figure 2. Photograph of Prototype Full Size Monolithic Ceramic Cross-Flow Filter Made With Blasch Process

Application

The application for this monolithic ceramic cross-flow filter product is to provide high temperature, high pressure removal of particulates from hot gas streams in coal fueled power systems. These power systems include IGCC (Integrated Gasification Combined Cycle) and PFBC (Pressurized Fluid Bed Combustor) types. The major benefit of the cross-flow filter design compared to the currently used candle filters is much higher filtration surface area. The monolithic cross-flow filter designed in this project has a filtration surface area of about 8.4 square feet vs. less than 3 square feet for a candle filter. This additional filter surface area is expected to yield significantly better filter efficiency.

In comparison to competitive box-shaped filters, there are two that are known. One is the "dead-end" ceramic membrane filter. This filter has high filtration surface area, but has so far been limited to dead-end dirty gas channels of relatively small cross-sectional size, that may be prone to plugging in application and also may be more difficult to clean via reverse pulse method. Although this project is not yet finished, the Blasch technology is already producing prototype monolithic ceramic cross-flow filters with dirty gas channels that are open on both ends and are approximately 8mm high. With a new mold, this same technology could be used to make channels of even larger size, if deemed beneficial. The other known ceramic cross-flow filter design is segmented, where grooved plates are stacked and adhered together to form the filter. The disadvantage of this filter design is that it has been known to delaminate/ separate in service. It is anticipated that the monolithic, one-piece design of the Blasch cross-flow filter will eliminate this delamination problem.

Future Activities

This Phase II SBIR project is approximately half finished. Future activities include further streamlining of processing techniques to produce ceramic cross-flow filters that are improved with regards to minimizing voids on internal filtering walls. Corrosion testing of composition #4-300 by Westinghouse will be completed. It also is planned that three prototype filters each of two compositions will be submitted to subcontractor Westinghouse for simulated filtration testing in their HTHP PFBC test vessel, followed by post-test characterization. Further specifics of this testing are included in the "Approach" section of this paper.

Acknowledgements

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