HIGH-TEMPERATURE GAS STREAM FILTER AND METHOD

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HIGH-TEMPERATURE GAS STREAM FILTER AND METHOD

BACKGROUND OF THE INVENTION

The present invention relates generally to the removal of solid particulate material from high-temperature gas streams, and more particularly the removal of such particulate material by employing a barrier filter formed of a carbon-carbon composite provided by a porous carbon fiber substrate with open interstitial regions between adjacently disposed carbon fibers selectively restricted by carbon integrally attached to the carbon fibers of the substrate. The United States Government has rights in this invention pursuant to the employer-employee relationship of the U. S. Department of Energy and the inventor.

High-temperature gas streams at temperatures frequently exceeding 800°F often contain solid particulate material which must be substantially, if not essentially entirely, removed from the gas stream before the high-temperature gas can be employed in the desired process or application. For example, the gasification of coal is utilized for producing high-temperature fuel gas at various pressures for use at temperatures greater than about 800°F in various processes and applications such as providing the motive fluid for gas turbines in electrical power generating systems. In order for hot, particulate-bearing gases including fuel gas to be utilized at temperatures greater than about 800°F without damaging equipment associated with the anticipated use or causing an environmental pollution concern, essentially all of the solid particulate material borne by the hot gas stream must be removed from the hot gas stream to result in the gas stream having a preselected minimal solid particulate loading. In a typical utilization of a particulate-bearing hot gas stream, the particulate loading of the gas stream after "cleaning" is normally less than about 50 ppm and with essentially no particulates larger than about 10 microns.
The removal of solid particulate material from high-temperature gas streams such as fuel gas streams is normally achieved by employing various types of mechanisms including cyclones, electrostatic precipitators, and/or barrier filters such as panels, bag filters, candle filters and the like. These particulate removal mechanisms provide satisfactory removal of solid particulate material from hot gas streams especially when the gas stream is at a temperature less than about 1,000°F. However, it has been found that as the temperature of the gas stream exceeds about 1,000°F, the basic physics involved in the removal of the solid particulates from the gas stream and the properties of material suitable for the construction of the particle removal mechanisms present problems which render difficult the effective removal of such solid particulates so as to provide sufficiently clean gas streams. In fact, when the temperature of the gas streams exceed about 1,500°F, even further and different problems are involved. The design and construction of the particulate removing mechanisms, especially the barrier filters used for filtering particulate material from such very high-temperature gas streams are often constructed of ceramic material components. This use of ceramic material introduces design problems due to its structural rigidity, brittleness, and thermal expansion characteristics since such properties of ceramic materials are frequently different than the materials with which they interface when used in barrier filter assemblies or devices. Also, with barrier-type filters containing ceramic materials, process abnormalities or upsets as well as the normal filter cleaning procedures, which usually utilize intermittent pneumatic back-flushing operations for removing accumulated filter cake, impose considerable load pulses on the barrier filter which must be compensated for in order to prevent damaging the barrier filter element itself. These problems with present high temperature barrier filters are exacerbated when the pressure of the gas stream exceeds atmospheric pressure up
to pressures of about 30 atmospheres. These and other problems associated with the
design and maintenance of the structural integrity of barrier filters useful for cleaning high-
temperature, high-pressure gas streams have not been adequately addressed.

SUMMARY OF THE INVENTION

Accordingly, it is a primary objective or aim of the present invention to provide a
barrier-type filter for effectively removing solid particulate material from gas streams at
temperatures greater than about 800° F while substantially minimizing or obviating the
aforementioned problems associated with previously known barrier-type filters utilized for
cleaning high-temperature gas streams.

Generally, the particulate filtration device of the present invention is utilized in
combination with a substantially oxygen-free, solid particulate-bearing gas stream at a
temperature greater than about 800° F. The filtration device comprises: carbon fibers
disposed in an array defining a substrate with open interstitial regions provided between
adjacently disposed carbon fibers that are of a size sufficient to provide the substrate with
throughgoing porosity; and, carbon integrally supported by the carbon fibers and
substantially filling the open interstitial regions between the adjacently disposed carbon
fibers to sufficiently restrict the size of pores defining the throughgoing porosity for
permitting relatively unrestricted passage of the gas stream therethrough while removing
therefrom substantially all of the solid particulate material that is over a preselected
particle size.

This carbon-carbon filter for removing particulate material of a particle size larger
than a preselected particle size from a gas stream at a temperature greater than about
800° F, is produced by the steps which comprise: providing a substrate of carbonaceous
fibers with pore-forming open interstitial regions between adjacently disposed fibers; and,
sufficiently filling these open interstitial regions with carbon integrally attached to and supported by the fibers for providing the interstitial regions with throughgoing passageways of a pore size sufficient to provide for the passage of the gas stream while preventing the passage of particulate material larger than a preselected particle size.

The barrier filter of the present invention is for use in cleaning high-temperature gas streams which are substantially free of oxygen such as characterized by fuel gas streams or so-called inert, i.e., oxygen deficient, gas streams. This requirement for using the filter of the present invention with gas streams substantially free of oxygen is due to the fact that at temperatures greater than about 1,000°F, oxygen in the gas stream reacts with and oxidizes the carbon in the filter so as to reduce the effectiveness or the structural integrity of the filter.

The open interstitial regions between adjacent carbon fibers in the substrate are preferably of a pore size substantially larger than about 10 microns, while the through-going porosity or passageways provided by the partial filling of the open interstitial regions with carbon integrally attached to the fibers are of any preselected size, preferably less than about 10 microns, and more preferably less than about 5 microns.

Other and further objects of the present invention will become obvious upon an understanding of the illustrative embodiments and method about to be described or will be indicated in the appended claims, and various advantages not referred to herein will occur to one skilled in the art upon employment of the invention in practice.

DETAILED DESCRIPTION OF THE INVENTION

As briefly described above, the present invention is directed to a barrier-type filter for removing solid particulate matter borne by substantially oxygen-free gas streams at temperatures greater than about 800°F and in the range of about 800°F to 3,000°F.
This filter is a carbon-carbon composite constructed of a porous substrate of multiple-ply carbon fibers with open interstitial regions between adjacently disposed carbon fibers that are selectively partially filled with a carbonized binder and/or deposited carbon which is integrally attached to the fibers as well as attaching the fibers to one another for providing the resulting filter with a high level of structural integrity and with a throughgoing porosity of a selected pore size, preferably, less than about 10 microns and more preferably, less than about 5 microns. With this filter construction, the hot gas stream can readily pass through the many pores defined in the filter between the adjacently disposed carbon fibers without undergoing an undesirable pressure drop while removing essentially all of the particulate material borne by the gas stream so as to provide a clean high-temperature gas stream with a selected solid particulate loading, preferably less than about 20 ppm, and preferably with no particulates greater than the selected size.

The porous carbon fiber substrate with open interstitial regions between adjacent fibers of a size substantially greater than about 10 microns can be produced in various configurations by employing any of several manufacturing techniques. One such substrate configuration is a cylindrical or tubular form produced by winding single or multiple-ply carbonaceous fibers, i.e., fibers of a carbon precursor such as rayon or cotton or, preferably carbon fibers previously prepared by the carbonization of such carbon precursors, about a suitably configured consumable or removable mandrel in any selected winding pattern for providing a multiple-layered winding of a tubular or cylindrical configuration. Such configurations may include cone-shaped structures as well as cylinders open at one or both ends.

These substrates are defined by carbonaceous fibers which are disposed in close proximity to one another with this positioning of the fibers being selectively variable so
as to provide open interstitial regions defined between adjacently disposed fibers of a
preselected size. These open interstitial regions substrates are greater than about 10
microns.

In order to bond the fibers to one another to provide a structurally stable winding
and to selectively tailor the size of the open interstitial regions between adjacently
disposed fibers in the winding, the carbonaceous fibers forming the winding are wet with
a carbonizable binder prior to, during, and/or subsequent to the formation of the winding
on the mandrel. This carbonizable binder also serves to sufficiently bond the fibers of the
winding together to permit handling of the green winding as, for example, if the
supporting mandrel is removed therefrom prior to final heat treatment. The wetting of,
these carbonaceous fibers during winding can be achieved by passing the carbonaceous
fibers through a bath of the carbonizable binder or by employing a wiper arrangement
wherein the fibers are wetted with the liqiuid binder.

The wetting of the carbonaceous fibers can also be provided by spraying or
painting the carbonizable binder onto the finished winding or during various stages of the
formation thereof. Selected or graduated penetration of the sprayed or painted
carbonizable binder into the fibrous substrate can be achieved by employing a perforated
mandrel and then subjecting the binder-wet fibers to a pressure loading or by employing
a vacuum inside the mandrel to draw the carbonizable binder into the various layers of
the winding. This painting or spraying of the liquid carbonizable binder onto the wound
substrate are preferred techniques for at least part of the wetting process since such
spraying or painting will produce a shell of relatively low permeability on exposed surface
regions of the substrate which defines the size of the open porosity at the surface of the
finished filter and thereby precludes deep penetration/retention of the solid particulate
material removed from the gas stream within the body of the filter so as to significantly reduce the tendency of the filter to undergo filter blinding.

Additional structural integrity may be achieved in the finished filter element winding by employing a perforated mandrel of a suitable relatively inert high-temperature metal such as titanium or niobium which will remain as a component of the finished filter element.

The carbonizable binders utilized in the fabrication of the filters of the present invention are resins which are capable of bonding together the individual carbon fibers in the substrate as well as providing for a relatively high yield of carbon when subjected to a suitable carbonization temperature as will be described below. When dried the binders also provide a degree of structural integrity to the filter element in its green state. Suitable carbonizable binders include thermosetting resins such as phenolics, epoxies, and furfuryl alcohols, or thermoplastic resins such as petroleum or coal tar pitch. Also, combinations of such resins may be satisfactorily used. The quantity of the carbonizable resin applied to the carbon fiber substrate during the formation thereof is the amount which will provide a tenacious bond between the fibers of the substrate for achieving a high level of structural integrity and which will provide the finished filter with a selected throughgoing porosity to provide for removing solid particulate material above a selected particle size from the hot gas stream passing through the filter.

After forming the green filter element with the carbonaceous fibers either in the form of carbon precursors or carbon and wet with the carbonizable resin binder, the green filter element is subjected to a temperature in the range of about 70°F to 500°F so as to drive off volatiles from the resin binder (and carbonizable fibers, if used) so that the resin may be sufficiently set to adequately bond the fibers together for permitting the
handling of the green filter element such as may be required for the separation thereof from the mandrel, if desired. After this drying step, the green filter element is heated in a vacuum, an inert atmosphere such as argon, or possibly hydrogen, to a suitable carbonizing temperature normally in the range of about 3,500 to 3,800°F for converting both the carbonaceous fibers, if used, and resin binder to carbon. The resulting carbon-carbon composite consists of essentially pure carbon, so as to possess very low chemical reactivity with the constituents of the high-temperature gas streams to be filtered. If a permanent mandrel is utilized for additional structural integrity purposes, it may present some chemical reactivity problems and care must be exercised when selecting mandrel materials so as to be consistent with both the filter processing steps and the constituents of the gas streams to be filtered.

As pointed out above, the winding of the carbon fibers results in open interstitial regions between adjacently disposed fibers. These open interstitial regions provide throughgoing porosity or passageways through the substrate at locations over the surface thereof between the adjacently disposed fibers which will provide for the passage of high-temperature gas stream and particulate material borne thereby that is of particle sizes which should be excluded from the gas stream before the use of the "clean" gas. In order to remove solid particulates of particle sizes larger than a selected size from the gas stream as it is passed through the filter, the size of these open pores or passageways in the winding can be initially provided for by several techniques, singly or in combination. Examples of techniques to adjust pore sizes include selecting the diameter of carbon fibers used in the winding with the smaller diameter carbon fibers providing smaller open interstitial regions between the carbon fibers, increasing the tension on the fibers during winding so that the fibers are disposed in closer proximity to one another,
increasing or decreasing the number of fiber layers or wall thickness of the fiber winding, and/or by using multiple layers provided by various winding patterns.

Upon completion of the winding or substrate, the final tailoring of the open or throughgoing porosity of the filter is achieved by the amount of the resin binder applied to the exterior of the substrate (e.g. the painting or spraying step mentioned earlier) and converted to carbon. However, a satisfactory alternative to selectively tailor the filter to provide the desired throughgoing porosity is to form a filter element with the fibers and the carbonizable binder wherein the porosity through the open interstitial regions is of a pore size larger than desired of the final filtering element. The tailoring of this partially finished filter element to selectively further reduce the size of these pores is then achieved by depositing carbon onto the surface of the carbonized binder such as by employing a conventional chemical vapor deposition (CVD) operation using a suitable carbon-bearing gas such as methane or carbon monoxide. In the chemical vapor deposition operation, the carbon-bearing gas atmosphere surrounding the substrate is suitably heated to convert carbon in the gas to pyrolytic carbon which is tenaciously deposited on the carbonized binder and fibers, especially in the interstitial regions of the carbon-carbon composite. This deposition operation is easily controlled so as to readily provide for selectively tailoring the filter porosity. Also, by using such a CVD pore size tailoring technique, the pyrolytic carbon is predominantly deposited in the near surface regions of the filter so as to substantially reduce the potential for filter blinding problems.

The barrier filter of the present invention can also be of a flat or arcuate shape as provided by using carbon fiber substrates wherein the single or multiple-ply carbonaceous fibers, preferably carbon, are placed in selected spatial relationships and orientations to one another to form mats or panels. However, if desired, the carbon-fiber substrate can
be formed from layed-down randomly oriented carbon fibers. Also, the carbon fibers can
be woven or interlaced to define a webbing-like structure. As with the substrate formed
by winding the carbon fibers, as described above, the carbon fiber panels in the form of
mats or webbings are wet with a selected amount of the carbonizable binder during or
subsequent to the formation of the substrates to bind the fibers together and for partially
filling the open interstitial regions between adjacently disposed fibers to achieve the
desired level of open or throughgoing porosity for the filter. These resins in the fibers as
well as the carbon fiber precursors are converted to carbon by employing the carboniza-
tion technique generally described above. Also, if desired, the final tailoring of the pore
size in the interstitial regions can be achieved by depositing additional carbon in the
interstitial regions by using a CVD operation.

If desired, several layers of the matings or webbings can be stacked together to
provide a relatively thick filter panel for additional structural integrity. Also, if desired, the
matings or webbings of the carbon fibers can be placed on or about a perforated free-
form of a high-temperature metal such as described above with this pre-form remaining
in place for adding structural integrity to the filter. The carbonizable binder will contact
this pre-form and bind it securely to the carbonaceous fibers to assure a structurally
strong filter element.

During the wetting of the carbonaceous-fiber substrates with the carbonizable
resin binder, additional amounts of the binder over that needed for controlling the open
porosity through the filter can be applied to selected surface regions of the substrate for
forming local high density zones so as to provide features such as high-strength bearing
shoulders, relatively impermeable end-caps, and surface regions which can be machined
or otherwise configured for filter mounting purposes. For example, in the fabrication of
cone-shaped filters, additional amounts of the carbonizable resin can be applied to the tip regions of the cone to close the pores at that end of the filter. Also, such additional carbonizable resin can be applied near the open end of the cone-shaped filter to provide a relatively dense region which can be machined to provide an interface for attachment of the filtering element to a filter mounting. In the case of the flat or curved-carbon filter panels formed of mats or webbings, additional carbonizable binder can be placed in peripheral regions of the filter for increasing the structural integrity of the filter, sealing the peripheral edges of the filter, and providing a surface for the mounting of the filter panel in a suitable filter support housing.

It will be seen that the carbon-carbon filter of the present invention is provided with open porosity, the pore size of which can be selectively tailored so as to prevent the passage therethrough of relatively solid particulate material down to a particle size of only about 5 microns. The carbon-carbon filter employing the carbon fibers therein provides for a structure which possesses high strength under both tension and compression loadings and yet has sufficient compliance for use in applications where a strain may be applied to the filter since the carbon-carbon filter of the present invention can undergo significant strain without fracturing. Also, the thermal expansion coefficient of the carbon-carbon filter is relatively low as compared to those of the ceramic filters employed for filtering particulate material from high-temperature gas streams so as to obviate the design problems associated with the use of ceramic filters in high temperature gas filtering applications. The carbon-carbon filter is formed of essentially pure carbon so as to be relatively resistant to corrosion and contamination and is compatible with constituents of most process gas streams, except for its reactivity with oxygen.
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ABSTRACT OF THE DISCLOSURE

A carbon-carbon barrier-type filter for removing solid particulate material from substantially oxygen-free, particulate-bearing gas streams at temperatures greater than about 800°F. The filter comprises a carbon fiber substrate of configuration selected for the filter and a carbonized resin binder which partially fills open interstitial regions between adjacently disposed carbon fibers in at least the near surface regions of the substrate to provide the resulting carbon-carbon composite with throughgoing porosity of a selected pore size. The final size of the pores defining the throughgoing porosity can also be tailored by depositing pyrolytic carbon on surface regions of the carbonized resin binder.