COMPACT HOTGAS CLEAN-UP SYSTEM FOR PARTICULATE REMOVAL
(HOTGASYS-1)

Johnny Marcher
Lars Tinggård Johannesen
Research Engineers
&
Hans Frost
Project co-ordinator

Contract No. JOF3-CT97-0047

PUBLISHABLE REPORT

Mar. 1, 1998 to Aug. 31, 2000

Research funded in part by
THE EUROPEAN COMMISSION
in the framework of the
Non Nuclear Energy Programme
JOULE III

COMPACT HOTGAS CLEAN-UP SYSTEM FOR PARTICULATE REMOVAL

HOTGASYS-1

ABSTRACT

Advanced power generation cycles from solid fuels can contribute significantly to future European energy security. This will require particle-free combustion / gasification product gases, at high tempera-tures, in order to achieve significant energy efficiency and environmental benefits.

State-of-the-art hot gas particle clean-up technology is typically based on ceramic candle filters (rigid and soft) and to a lesser extent on smaller, one-piece honeycomb cross-flow filters. The technology is still in a distance from providing reliable and cost-effective particulate removal from hot gases. Major problem areas involves mechanical failure due to ash particle bridging, filter thermal stability and materials compatibility issues.

Assembling many individual filter elements (candles) into one large filter unit, are to be viewed as *a giant honeycomb*. All one-piece honeycomb structures have the advantage of an extremely high mechanical strength. One-piece honeycomb structures are however rarely produced with diameters of more than 500 mm. By enclosing many separate elements in a vessel or container, it becomes possible to obtain the same behavior as that of a one piece fabricated cross-flow honeycomb structure.
It is expected that the unit described hereby can make more efficient use of the volume taken up by filtering systems using appropriate packaging technology. The weight reduction is important for any system where load transients are common, such as emergency and peak-power electrical plants.

The objective of the project was then to develop a compact hot gas particulate clean-up system by integrating three specifically tailored, advanced high temperature ceramic materials (Silicon Carbide, high strength Cordierite and Aluminum Titanate) into a novel, and cost-effective filter design with the following objectives:

1. To overcome problems with current candle technology in terms of mechanical failure due to ash bridging, resulting in filter system breakdown and costly down-time and service.
2. To improve filter thermal stability and materials compatibility.
3. To increase the filter surface to volume ratio with simultaneous reduction of overall filter weight resulting in a very compact unit.
4. To provide high filtration efficiency - low pressure drop filter media at a lower manufacturing cost.

Discussion:

The design of tubes assembled into a giant honeycomb proved to be a feasible solution. This design alone is responsible for overcoming several problems from known ceramic filters for hot gas applications:

Ash bridging does no accour in the gap between the filterelements in cold conditions.
No thermal cracks over the unit structure’s cross section is possible.
Elimination of mechanical stress transferred to neighbouring elements.
Large scale manufacturing is made possible.
The filter surface is increased compared to other filters at same volume and the unit is therefore very compact.

Further more the design of the filter with floating support beams for the elements makes it possible to vary the size of the filters to customer demand, to a certain extend.

One of the greater problems were to design an effective sealing of a honeycomb made from individual tubes, mainly due to the manufacturing tolerances of the ceramics, and the differences in the thermal expansion coefficient of the filter elements and the vessel.

Corrosion maps has been evaluated for the three ceramics, focusing on the reactions in coal combustion and gasification environments.
All three ceramics proved to posses the material specifications required to provide high filtration efficiency and a low pressure drop.

The cold flow test showed filtration efficiencies in the range of 99,90 % for both SiC and Cordierite, and apparent permeabilities in the range of $2{,}76 \times 10^{-12}$ m² for SiC. Further more the test showed that SiC and Cordierite has got the mechanical strength needed, making the filter less sensitive to load transients.

Final conclusion

A compact hotgas filter with the desired specifications has been designed.
The three ceramic materials have been developed, tested, refined and evaluated.
A full-scale prototype has been constructed and cold flow tests have been carried out for two of the materials, and have proven that such a filter works under these conditions.

Table of contents:

PARTNER PRESENTATION

Organization name	Organization activity	R&D function and role in the project
NOTOX A/S Baldersbuen 20 2640 Hedehusene Denmark Phone +45 70 23 89 89 Contact person: Johnny Marcher	Production of combustion aftertreatment and hot gas clean-up systems (particulate traps and catalytic converters).	Developer and manufacturer of compact hot gas filter. Manufacturer of non-clay bonded SiC filter elements and membrane coatings. **Coordinator,** **Partner 1**
CERTH/CPERI 6th Km Charilaou-Thermi Road P.O. Box 361 57001 Thermi Thessaloniki Greece Phone +30 31 498192 Contact person: Dr Athanasios Konstandopoulos	Research in particle technology and combustion aerosols for energy and environmental applications.	R&D performer on lab scale particle flow and filtration testing, computer modelling and system engineering. **Partner 2**
ABBCAR Utility Power Plants Division of ABB Stal AB Sweden - 612 82 Finsprong	Manufacturer of PFBC and fossil fuel combustion systems.	Industrial advisor during the first phase of the project and prospective end user of the developed filter system during the anticipated second phase (Demonstration) **Partner 3**
CERECO 72nd Km of Athens-Lamia National road P.O. Box 146 34100 Chalkida Greece Contact person: Dr Anna Tsoga	Industrial services in the field of ceramics and refractories.	Developer and manufacturer of aluminum titanate filter elements. R&D performer on membrane deposition and materials characterization. **Partner 4**
CTI (Ceramiques Techniques et Industrielles SA) La Resclause Route de Saint Privat - BP 34 F-30340 Salindres France Contact person: Jean Pierre Joulin	R&D and production of membranes and supports for tangential and frontal filtration, purification of gases and liquids.	Developer and manufacturer of cordierite elements and controlled pore size membrane coatings. Upscaling of aluminum titanate manufacturing. **Partner 5**
IK-DTU - Department of Chemistry on Technical University of Denmark DK-2800 Lyngby Denmark	Research in processing and environmental testing of technical ceramic porous materials.	R&D performer on design, computer modeling and materials characterization. **Partner 6**

OBJECTIVES

The objective of the proposed research is to develop a compact hot gas particulate clean-up system by integrating three specifically tailored, advanced high temperature ceramic materials (Silicon Carbide, high strength Cordierite and Aluminum Titanate) into a novel, and cost-effective filter design with the following objectives:

- To overcome problems with current candle technology in terms of mechanical failure due to ash bridging. Resulting in filter system breakdown and costly downtime.
- To improve filter thermal stability and materials compatibility.
- To increase the filter surface to volume ratio with simultaneous reduction of overall filter weight resulting in a very compact unit.
- To provide high filtration efficiency - low pressure drop filter media with a lower manufacturing cost.

By assembling many individual filter elements (tubes) into one or several large structures the proposed rigid, low density filter units are to be viewed as giant honeycomb structures. One-piece honeycomb structures are rarely produced with diameters of more than 500 mm. All one-piece honeycomb structures have the advantage of an extremely high mechanical strength. By enclosing many separate elements in a vessel or container, it becomes possible to obtain the same behavior as that of a one piece fabricated cross-flow honeycomb structure. All mechanical forces are transferred correctly through the walls to the canning container.
It is expected that the unit described hereby can make more efficient use of the volume taken up by filtering systems using appropriate packaging technology. The weight reduction is important for any system where load transients are common, such as emergency and peak-power electrical plants.

The system developed should have the following characteristics compared to a similarly sized candle-based system:

- Considerable increase in mechanical strength, >25 MPa
- Higher operating temperature >1.000°C
- Considerably lower pressure drop with filtration efficiency as high as 99.9%
- Half the system weight for the same volume
- Doubling the filtration surface to volume ratio
- Considerably reduced system price
- No foreign patent dominance

TECHNICAL DESCRIPTION

Workpackage 1: Preparatory and supporting activities

Computer aided design and modelling of the hot gas filter were employed to elaborate and clarify fundamental design questions. Necessary filter production and testing equipment were custom designed and engineered and appropriate starting materials were procured and/or produced in the beginning and also in parallel with small-scale filter element development.

Workpackage 2: Small-scale elements manufacturing

Porous ceramic structures of simple shape were prepared from the different ceramic materials (non-clay bonded SiC, Cordierite and Aluminum Titanate) from ready-made powders or via solid state reactions (WP1).
The finished products showed clearly defined values for the specific surface, the pore size distribution and the pore volume. Thus, a very important parameter for the production of honeycomb structures is the particle size distribution of the starting powders as it influences the final pore size distribution of the ceramic piece.

Therefore during this work, different qualities of starting raw materials selected or produced in Workpakage 1 were investigated. X-ray diffraction analysis, laser particle size distribution measurements, pore size distribution measurements, (N_2 porosimeter), BET surface area calculations and SEM observations were used to characterise these powders.
Homogeneous mixing of raw materials, extrusion of small pieces with simple shapes (disks, rods and bars) and firing under various conditions followed. This part of the investigation included the sintering studies, which identified the firing conditions and the temperature range required for the synthesis of the appropriate stable phases. Ceramic pieces were characterised in terms of their structural, physicochemical and mechanical properties. These characterisations were combined with hot-stage X-ray diffraction analysis and thermogravimetric and differential thermal analyses in order to reveal the reaction mechanisms.
A systematic study of the processing parameters for the construction of honeycomb structure filter elements by extrusion from the raw materials identified was carried out. Parameters studied included the effect of various binders on the plasticity, formability and coherence of the shapes. Conditions for the production of easy-to-handle, plastic mass for each particular material and composition, were identified (effects of different binders and plasticizers, water to solids ratio, etc.). The mixing and homogenising conditions as well as the extrusion parameters (pressure, extrusion velocity, and temperature) were investigated. The effect of all these parameters on the final ceramic properties was also studied.
Drying of the extruded elements was another important step of the processing and were carefully studied and optimised. Finally, the appropriate firing cycles for the production of the desired refractory phase were determined for each case.

The outcome of this part of the work was the preparation of small size (60 mm in diameter) filter elements of flat disks. Also honeycomb shapes were manufactured for subsequent lab testing.

Slip-casting development of all three materials (SiC, Cordierite and Aluminum Titanate) into shapes of wedges acting as “channel closing” blocks and supports for the tubes took place.
In parallel, membrane deposition studies were carried out in order to fabricate the final filter elements. Dip-coating or airbrush-coating techniques was used for deposition. Solids content, pH and slip temperature, particle size distribution of the initial powders were factors that influenced the whole process and they were studied. The effect of the different preparation parameters on the properties of the membrane like the distribution, the thickness of the layers, the porosity and the thermal stability were studied.

Workpackage 3: Lab Scale Testing and Characterisation

Materials characterisations were performed just after preparation and after filtration and flow testing. This included physicochemical, structural and thermomechanical properties evaluation as well as corrosion resistance measurements. Flow and filtration testing included initial experiments on filter coupons, while the evaluation of the small size filters units under simulated hot gas conditions followed.

Workpackage 4: “Full scale” element manufacturing

Filter elements of all three materials (SiC, Cordierite and Aluminum Titanate) were extruded. The processing conditions identified in WP2 for each material were employed. Final refinements of the conditions for the large-scale manufacturing were done. Special attention were paid to extrusion and drying condition optimisation to insure straightness, avoid crack formations and in every means keep dimensional stability.
Slip-casting of all three materials (SiC, Cordierite and Aluminum Titanate) into wedge shaped “channel closing” blocks acting as mechanical supports were done.
Membrane deposition, processing parameters identified in WP2, were also refined and optimised.
The outcome of this work-package was the optimisation of all processing parameters for manufacturing demonstration full-scale ceramic filter elements.

Workpackage 5: Filtration system engineering

This work-package involved the production a prototype filter system incorporating the filter elements produced in WP4 into a honeycomb unit and arranged in a vessel featuring an inlet and an outlet port.
Vessel steel material selection was adjusted accordingly to the number of vessels. Steels with high corrosion resistance were used for vessel production.
The system also integrated a filter-cleaning device. Subsequently the complete filter unit was tested with a large cold-air ventilator and a dust loaded fluid for visual test of the dust cake build up and function of the cleaning system. Operational and control strategies for system cleaning were undertaken combining computer modelling (WP1) and the experimental data of WP3.
Finally a techno economic assessment of each proposed solution were undertaken in order to quantify the real potential of the new system.

RESULTS AND CONCLUSIONS

Results obtained by and belongin to CERECO:

1. Synthesis of aluminum titanate based composites

Aluminum titanate (tielite) is known to exhibit a low thermal expansion coefficient (lower than that of fused silica) and a higher refractoriness than cordierite (melting point higher than 1800^oC). Its apparent low bulk expansion is mainly attributed to the presence of microcracks resulting from differential stresses between grains during cool down, due to a high degree of thermal expansion anisotropy. This property indicates aluminum titanate as a potential material for high-temperature structural applications. On the other hand the microcrack network being developed is responsible for the poor mechanical strength of tielite. This problem together with the thermal instability observed at temperatures between 850 to 1250^oC, remain the main limitations for a wider use of the material. CERECO succeeded in the framework of the project to face both of the above material problems by synthesizing tielite-mullite based ceramic composite.

2. Tailored made powder through spray drying.

Spray drying involves the atomization of a ceramic fluid feedstock into sprays of droplets, which are dried to individual powder particles in contact with hot air. It is one of the few continuous drying techniques that incorporate a particle formation process. The main parameters that affect particle size distribution of the produced granular material are the viscosity of the slurry, the flow rate of the slurry and the air and the type and the diameter of the nozzle. In the framework of the project CERECO studied the above parameters and optimized the operation conditions of the spray dryer thus obtaining the required particle size distribution of the granules without reducing significantly the efficiency of the process.

3. Technology for porous ceramics fabrication with controlled characteristics:

Tielite-mullite filter elements with 40% porosity and mean pore size of 20-25µm were developed and can be further tailored to specific applications.

Results obtained by and belongin to NoTox A/S:

1. Concept of assembling a monolith from individual filter channels.

By assembling many individual filter elements (channels) into a large structure, a rigid, low density filter unit to be viewed as a giant honeycomb structure is made. By enclosing many separate elements in a vessel or container, it becomes possible to obtain similar mechanical behavior as that of a one piece fabricated cross-flow honeycomb structure. All mechanical forces are transferred through the walls to the canning container. Thereby a very durable, and much more compact filter, having a higher effective filter area/system volume can be obtained, reducing the system weight accordingly.

Ceramic filter materials are preferred when the process environment contains hot gases, in particular with a low oxygen content and the possibility of a high sulfur content. One important feature of the system is the possibility for a built-in catalytic conversion arrangement. This simultaneous particle filtering and gas pollutants conversion technique in one container would lower the total installation volume, the cost and heat losses considerably.

2. Design and prototype of a filter unit comprising a filter monolith assembled from individual filter channels.

By assembling many individual filter elements (channels) into a large structure, a rigid, low density filter unit to be viewed as a giant honeycomb structure has been made. In order to obtain the desired advantages, in terms of filtration performance and durability, it is crucial to obtain a design that among other factors accomodates thermal expansion of the individual filter segments, without compromising the critical sealing at the ends of the filter channels. During the Hotgasys-1 project a large number of methods and technical solutions has been developed by NoTox AS, to overcome the technical challenges.
The final prototype manufactured by NoTox has proven to be operational, and fullfilling the original project goals.

3. Design of thin walled filter tube, development of production equipment and determination of production parameters.

Previously obtained (background) knowledge from the field of Diesel Filters and Hot Gas candle filters, has been used in the design of the individual filter segments, including the design of production equipment and in the determination of production parameters. To obtain virtually straight tubes/single channels complying with the extremely tight dimensional tolerances required for the assembly of a monolith from single channels, a thorough optimisation of both the flow paths in the extrusion dies and of the rheological properties of the ceramic paste has been performed. Besides the composition of the ceramic paste has been optimised in terms of material strength. The micro porous membrane has been improved compared to background knowledge in terms of homogeneity and by reducing membrane entrainment in the underlaying coarse substrate.

Results obtained by and belongin to CERTH CPERI:

1. Computational design and simulation of compact hot gas filters and their cleaning system by back pulsing

The flow and filtration behavior of a novel hot gas cleaning system during normal operation and reverse pulse cleaning has been simulated computationally for the operating conditions, using 3D, transient calculations. The simulation concerns ceramic filter elements assembled in a specific structure. Further development is required to extend the application to other filter element and system designs. The computational methodology provides a useful tool for developing new filtration systems and defining the optimum operating parameters. This could be useful in applications like advanced cycle power production, biomass/waste incinerators, marine diesel engines, heavy fuel boilers, recovery of catalyst fines in FCC units, combined gas/particle removal with catalyzed elements and cement production.

2. Methodology and experimental facility for testing hot gas coupon and candle filters.

An experimental facility, which can expose filter sample materials to particle laden flows at temperatures up to 1000^{o}C and pressures up to 10 bar has been constructed [1]. The filter testing flow rig is instrumented with particle measuring instrumentation, which provides continuous, on-line measurement of the particle size, concentration and velocity. The main advantage of the experimental facility is that the filter can be tested at realistic conditions of temperature, pressure and face velocity, providing thus evaluation services of filter materials to filtration system manufacturers and end-users.

3. New non-destructive, high temperature, flow-based technique for membrane structure characterization

A new technique was developed for characterizing microporous structures in a non-destructive way at high temperatures. The technique provides a means for determining the effective pore size of ceramic membranes, in-situ, regardless of the size or shape of the sample.

Results obtained by and belongin to Céramiques Techniques & Industrielles s.a

1. The determination of production parameters regarding Cordierite.

To obtain virtually straight tubes/single channels complying with the tight dimensional tolerances required for the assembly of a monolith from single channels, a thorough optimisation of the prodution parameters were done.

2. The determination of production parameters regarding Cordierite.

To obtain virtually straight tubes/single channels complying with the tight dimensional tolerances required for the assembly of a monolith from single channels, a thorough optimisation of the prodution parameters were done.

ECONOMIC, INDUSTRIAL, SOCIAL and ENVIRONMENTAL IMPACTS

Direct Industrial Applications

The present research aimed at developing a compact, more efficient than competing candle technology hot gas clean-up system for particulate removal by integrating novel design aspects and manufacturing methods employing advanced ceramic materials of improved thermomechanical characteristics.

Highly efficient ceramic particulate filters are required in a wide variety of hot gas clean-up applications including such processes as:

- coal combustion (pressurized, atmospheric, and circulating fluidized bed)
- coal gasification
- incineration (radioactive, pathological, mixed wastes, etc)
- catalyst recovery and processing
- chemical and petrochemical processing.

In coal combustion and gasification processes, ceramic filters are used to provide a high degree of particulate removal at high temperatures, in order to protect process equipment (such as a gas turbine) downstream of the combustor/gasifier. Filter effluent gas must conform to the limits on allowable particulate as specified by the turbine manufacturer.

PFBC (Pressurized Fluidized Bed Combustion) is a modern technology for clean power generation from coal featuring low emissions, low fuel consumption (lowest possible fuel cost), compact design (enabling the utilization of use old sites) and fuel flexibility.
The combination of these features and their associated benefits makes PFBC a very attractive proposition to most of the world’s utilities. In the area leading companies are ABB Carbon, Westinghouse and Babcock that have been performing R&D for several years.
In modern PFBC plants, the overall thermal efficiency can be increased by connecting a gas turbine directly on the combustion chamber outlet. Until now, this has proven very difficult, as the gas containing large amounts of solids erodes or wears out the gas turbine parts within an extremely short period of time. It is obvious that the integration of an efficient and reliable hot gas filter in the in the high temperature/high pressure PFBC system environment is a non-trivial matter of great importance. Thus, the development of a highly efficient, compact filter with improved thermo mechanical and corrosion resistance characteristics with respect to current technology, will be a highly welcomed development will lead to very important benefits on the total economy of the PFBC power plant.

Typical examples of gasification units incorporating ceramic filters are the Dutch Shell plant in the Netherlands for coal gasification and the Puertollano ICCG plant in Spain, which both use a Schumacher candle filter system. Ansaldo in Italy is also involved in gasification plants using wood and waste for power production.

The process industries are potential customers for using ceramic filters to remove fine particles (often submicron) in gas phase separations such as:

- Catalyst/precious material recovery applications.
- Catalytic cracking/refining industry applications which include recovery of the catalyst which is circulating in a fluidized bed under pressure at temperatures over 500 ^{o}C for coke oxidation (regeneration) and recovery of the catalysts in de-sulfurization processes.
- Chemical Process Industries for environmental protection in plants for production of sulfur, titanium oxide and nitric acid.
- Calcination processes and metallurgic operations
- Cement Processing

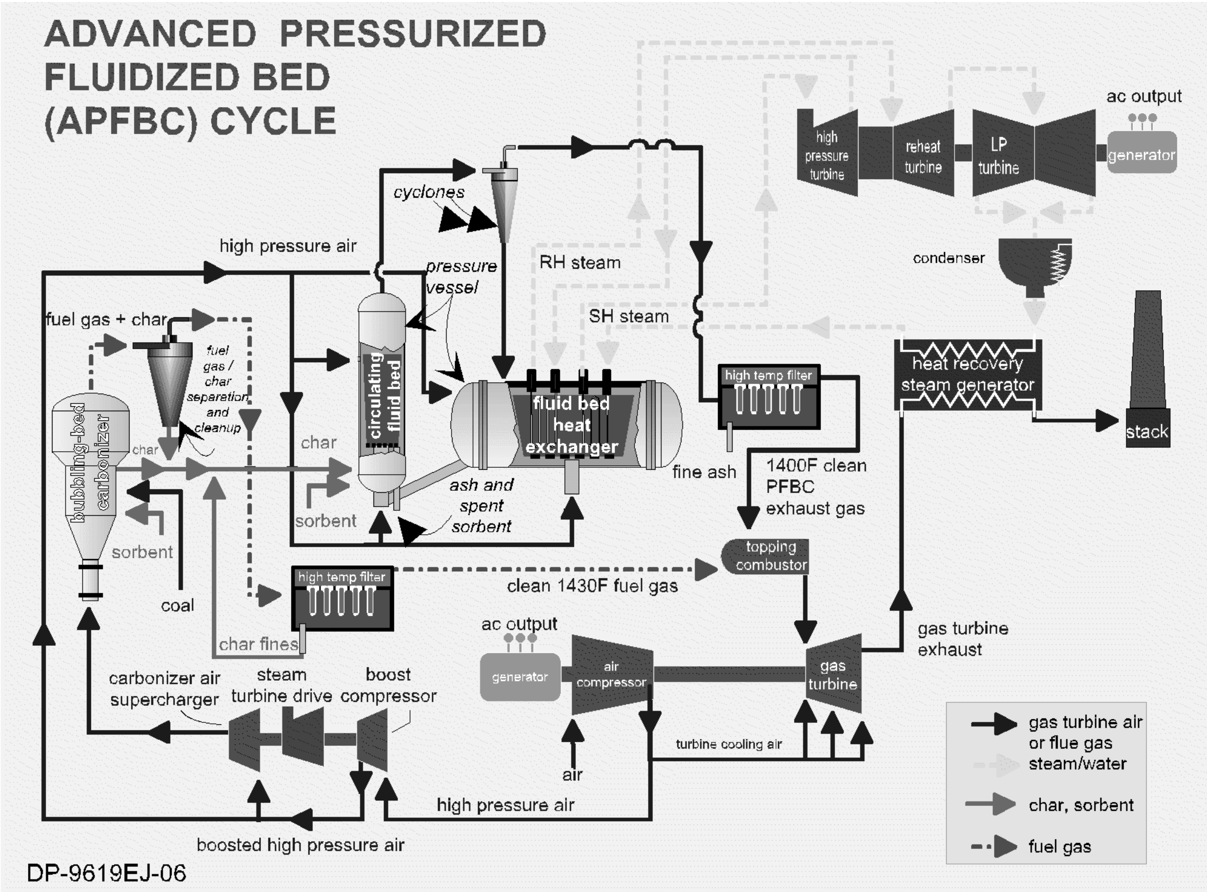
In incineration applications, ceramic filters are used to clean particulate emissions to levels conforming to environmental standards. In most waste incineration plants conventional baghouse technology is involved (temperatures less than 200^{o}C), but some plants have since the late 1980’s been changing to ceramic filters for eliminating the emissions by hot gas cleaning. Also hospital waste incineration at high temperatures is a promising application area.
Thus, with respect to the potential market, various groups of users take shape: power stations, industrial plants, waste incineration plants, etc. Industrial sectors that will immediately benefit from the successful results of the project are mainly those dealing with the manufacturing of PFBC and coal gasification systems for increasing energy efficiency.

Indirect Industrial Applications

The development and the optimization of novel technologies for the production of porous ceramic structures and membranes will not only be valuable in the hot gas clean-up filter manufacturing field but also in all areas where these kind of products find applications:

- ceramic membranes and liquid filtration domain
- advanced ceramics areas (ceramic parts with improved mechanical properties)

Potential application of the project



References

1. A.G. Konstandopoulos et al., “The Design of a Novel Hot Gas Filter Unit”, Fourth International Symposium on Gas Cleaning at High Temperatures, University of Karlsruhe, 1999.
2. U.S. Department Of Energy website http://www.energy.gov/