CARBON COMPOSITE FILTER FOR RADIOACTIVE WASTE STORAGE CONTAINERS

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INTRODUCTION

Transuranic (TRU) waste containers currently used for shipping and storing waste materials are required to withstand 7.3 psi internal pressure according to Department of Transportation (DOT) Type "A" specifications. Due to excessive cost of containers capable of withstanding such pressure buildup, containers not being transported by air have been exempted from this requirement. Thus, two problems exist relative to present TRU waste containers: (1) containers exempt from the pressure requirement have the potential for leaking radioactive contaminated waste; and, (2) containers which are truly gas-tight and designed to contain a certain amount of pressure can become potentially hazardous in the event of overpressure caused by gas generation.

Gas generation studies conducted at Rocky Flats revealed that many plutonium contaminated waste materials decompose into gaseous products because of exposure to alpha radiaiton. The gases generated, consisting largely of hydrogen, are generated at the rate of 1 to 16 cubic centimeters per day over an extended period of time. Generation of gaseous products in containers used in shipping and storage of TRU waste presents a potential release hazard in the event of container rupture due to pressure buildup. Studies designed to identify, control and/or neutralize materials responsible for gas generation are being conducted with limited success. Containers capable of withstanding a large pressure buildup have been designed and evaluated, but are very heavy and costly. Thus, the need for an inexpensive, high efficiency particulate filter capable of retaining contaminated particles while venting the generated gaseous products was investigated as a possible solution to this problem.

Commercially available filtering materials have been evaluated and found unsatisfactory with regard to this particular application. However, a low density ($0.3\ g/c^3$) carbon-carbon composite, previously developed for use as a high temperature thermal insulation, has been evaluated for use as a high efficiency particulate filter. This material which exhibits a combination of high porosity (~90 vol %) and large surface area was observed to exhibit excellent particulate removing and air flow characteristics. Several filters consisting of a carbon-carbon plug (19 mm diameter by 12.7 mm thick) were fitted into a drum plug and held in place with RTV 116 high temperature silicone sealant. Smoke testing using dioctylphthalate (DOP) showed these filter elements to be >99.0% efficient in filtering out particles 0.3 μm in size, or greater. Excellent filtering properties, together with good mechanical and thermal properties resulted in further evaluation of this material for use as a nuclear grade filter.

EXPERIMENTAL

Fabrication

Low density carbon bonded carbon fiber composites were fabricated by vacuum molding using a dilute water slurry of carbon fibers and an insoluble particulate phenolic binder. A perforated metal mandrel was used to form composites of the desired shape. After molding, the parts were removed from the mandrel and dried at 60 °C for several hours, then heated to 130 °C which caused the resin to flow and cross link. The semirigid structure was then subjected to a temperature of 1350 °C which pyrolyzed the polymeric binder to produce a carbon bonded fibrous structure. The carbon fibers used in this study were obtained by pyrolyzing rayon fibers at 1350 °C. Pyrolysis of the rayon yielded about 40 wt % carbon with about a 15-20% linear shrinkage. Following the carbonization cycle, the fibers were drawn through a Wiley mill in order to deagglomerate bonded fibers generated by either the chopping or pyrolyzing operations.

Porosity and surface area of the composite material, which dictate filtering efficiency and air flow characteristics, are directly related to the composite's density. The density of the carbon-carbon composite is primarily dependent on fiber length, fiber concentration, and fiber to resin ratio. Thus, a wide range of filtering characteristics (efficiency, air flow resistance, gas adsorption) and mechanical properties can be obtained by varying the carbonaceous composition and processing parameters.

Testing

Evaluation of the low density carbon-carbon composite for use as a high efficiency particulate air filter was conducted by smoke testing using dioctylphthalate (DOP) particles (~0.3 μm) according to Military Specification F0051079-D. The material evaluated had a density of 0.22-0.24 g/cc, compressive strength of about 170 psi, and porosity in the range of 85-90%.

Filter test units were constructed by inserting a 19 mm diameter by 12.7 mm thick piece of carbon into a drum plug held in place with a high temperature silicone sealant and a TAC welded perforated sheetmetal disk. A cross-sectional view of a filter unit is shown in Fig. 1.

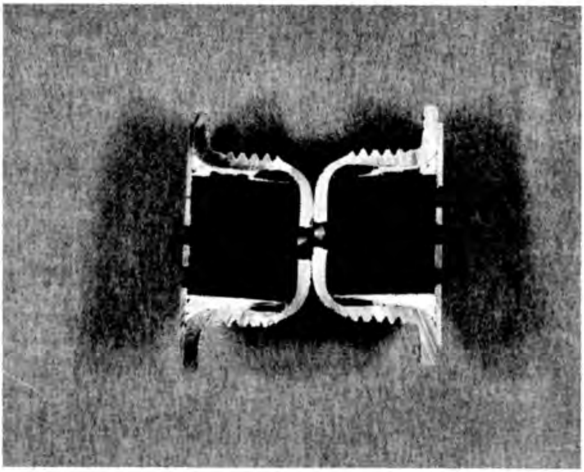

Figure 1. Cross-sectioned Filter Unit

DOP testing of the filter was performed by the Rocky Flats filter certification laboratory. Testing was conducted after mounting the filter element on a 55-gallon drum and forcing DOP particles into the drum using a cold aerosol generator equipped with a compressor. The quantity of particles escaping through the filter was measured using a portable ATI photoameter. Filtering efficiencies of several test filters were determined at 1, 2, and 5 pounds of air pressure corresponding to air flows of 3-11 Standard Liters per Minute (SLM).

Results

A total of five test specimens were assembled for evaluation. These five specimens are similar in density and have the fibers oriented transverse to the air flow. The specimens were DOP tested at 1, 2, and 5 psi corresponsing to air flows of about 3.3, 5.2, and 11.2 SLM, respectively. The specimens were tested before and after exposure to vibration (1,000,000 cycles at a 0.015-inch stroke), shock impact (consisting of a 4-foot free fall onto an unyielding surface, repeated ten times), and exposure to 55 and 246 ^{o}C for 24 hours, and -37 ^{o}C for 48 hours.

Results of DOP testing at 1, 2, and 5 psi are shown in Table I. Filtering efficiencies ranging from 99.5 to 99.85% were obtained when tested at 1 psi. These efficiencies improve when tested at 2 and 5 psi to give a range of 99.84 to 99.95% at 5 psi. This improvement in performance has been attributed to compression of the silicone sealant used in bonding the carbon plug to the metal holder.

TABLE I

PERCENT FILTERING EFFICIENCY AS A FUNCTION OF RESISTANCE PRESSURE

Resistance (psi)	Air Flow (SLM)	Sample Number 1	2	3	4	5
1	3.3	99.54	99.81	99.39	99.85	99.72
2	5.2	99.74	99.80	99.56	99.87	99.80
5	11.2	99.91	99.95	99.84	99.94	99.93

The same five specimens were DOP tested again at 5 psi after exposure to vibration, impact, 55 and 246 ^{o}C for 24 hours, and -37 ^{o}C for 48 hours with results shown in Table II. Filtering efficiencies of these five specimens were not affected by any of the induced stresses, with final efficiencies ranging from 99.87 to 99.94%.

TABLE II

PERCENT FILTERING EFFICIENCY AT 11.2 SLM
(5 PSI RESISTANCE)

Condition	Sample Number 1	2	3	4	5	Average
As Fabricated	99.96	99.88	99.96	99.94	99.96	99.94
After Vibration	99.84	99.88	99.80	99.90	99.91	99.87
After Shock Impact	99.90	99.98	99.80	99.98	99.90	99.91
After 55 ^{o}C/24 hr	99.82	99.97	99.80	99.92	99.98	99.90
After 246 ^{o}C/24 hr	99.97	99.98	99.88	99.92	99.90	99.93
After -37 ^{o}C/48 hr	99.97	99.99	99.80	99.97	99.90	99.93
AVERAGE	99.91	99.95	99.84	99.94	99.93	

CONCLUSIONS

Rupture and/or bulging of TRU waste containers experiencing pressure buildup due to gas generation can be safely and inexpensively eliminated by fitting each container with a nuclear grade filter vent. The carbon composite filter evaluated in this study meets all material and filtering requirements and is strongly recommended for use as a filter vent in waste containers.