

STRENGTH ENHANCEMENT AND APPLICATION DEVELOPMENT OF CARBON FOAM FOR THERMAL MANAGEMENT SYSTEMS

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ABSTRACT

Carbon foam is recognized as having the great potential to replacement for metal fins in thermal management systems such as heat exchangers, space radiators, and thermal protection systems. An initial barrier to implementation was the inherent weakness and friability of the carbon foams. Ceramic Composites Inc. has demonstrated the ability to increase the compressive strength by 2½ times through the treatment of the carbon foam ligaments with a uniform silicon carbide coating, serving to enhance strength and reduce friability, with minimal influence upon the thermal properties. The process is amenable to automation using commercially available equipment. An overview of the technical approach will be presented, along with the envelope of enhanced material properties achieved under the program.

INTRODUCTION

Carbon foams of various pedigrees have been studied since the late 1960s when the first reticulated foams were produced.ⁱ These foams were thermally insulating and remained so for many years with research in the 1970s focused primarily upon producing carbon foams from alternative precursors. In the early 1990's, researchers at the Wright Patterson Air Force Base Materials Lab pioneered mesophase pitch derived graphitic foams, specifically for replacing expensive 3-D woven fiber performs in polymer composites and as replacements for honeycomb materials. This was one of the first high thermal conductivity carbon foams and the technology was licensed by MER Corporation of Tucson, AZ. Later, West Virginia University developed a method that used coal as a precursor for high strength foams with excellent thermal insulation properties, a process licensed by Touchstone Research Laboratories of Triadelphia, WV. Ultramet produces a similar product of thermally insulating pyrolytic CVD carbon on reticulated vitreous carbon cells.ⁱⁱ In 1997, James Klett at the Oak Ridge National Laboratory (ORNL) reported the first graphitic foams with bulk thermal conductivities greater than 40 W/m·K (Figure 1). This process was licensed to POCO Thermal Materials of Decatur, TX and the product trade named POCO Foam. POCO later developed POCO HTC, which has higher thermal conductivity and density. The POCO materials are made in sheets, similar to cakes, with properties in the z-direction radically different than in the x-y directions. The process for producing POCO Foam is illustrated in Figure 2.

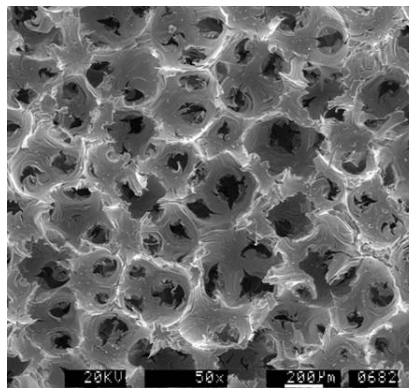


Figure 1: Micrograph of ORNL carbon foam.

These recent carbon foams are of interest due to their high thermal conductivity (primarily in the z-direction), low density, and open cell structure. With thermal conductivities similar to aluminum, they are under consideration for thermal protection systems, thermal management systems (both active and passive), solar radiators, industrial heat exchangers, electronics cooling, and noise absorption. In electronic systems, the use of a

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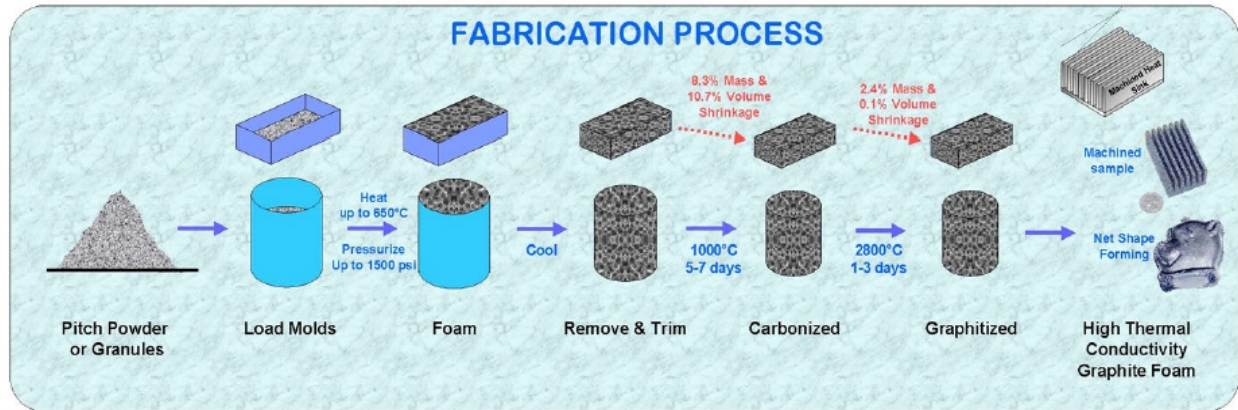


Figure 2: Fabrication process for POCO Foam.



Figure 3: Automotive radiators which demonstrated 33 to 250% performance improvement.

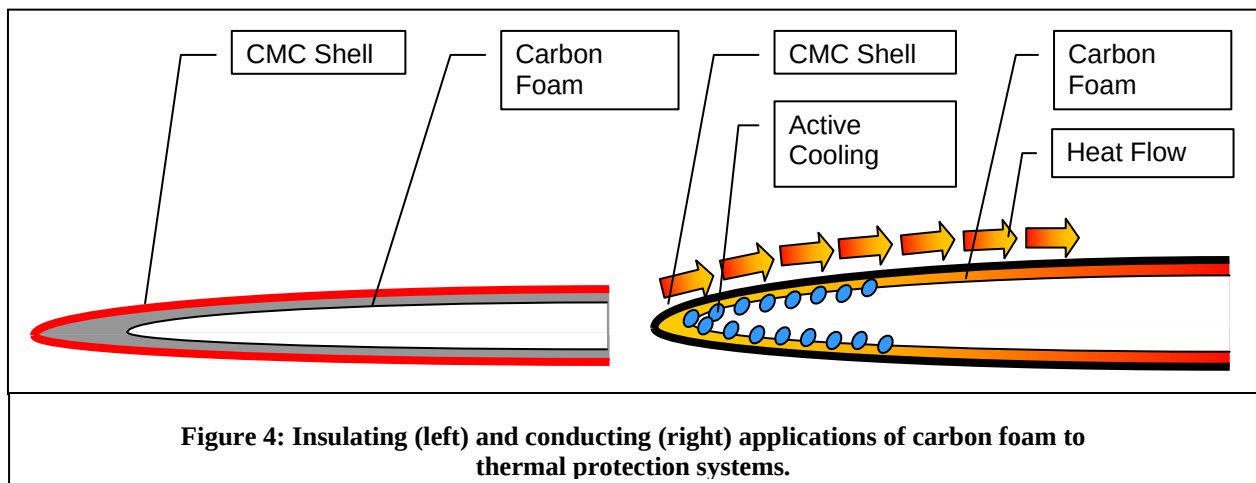


Figure 4: Insulating (left) and conducting (right) applications of carbon foam to thermal protection systems.

carbon foam heat sink as a direct replacement for an aluminum heat sink lowered the chip temperature by 6°F.ⁱⁱⁱ In automotive applications the performance of equivalent sized radiators (Figure 3) was increased 33 to 250%.^{iv} In Thermal Protection Systems (TPS) there are two approaches under consideration for utilizing carbon foams. Allcomp Inc.^v, Materials and Electrochemical Research^{vi}, Touchstone^{vii}, Ultramet^{viii}, Fiber Materials Incorporated^{ix} have suggested placing *insulating* carbon foam below a Ceramic Matrix Composite (CMC) shell to protect the craft interior from the heat of re-entry. This is shown in the left view of Figure 4. Thompson^x of San Diego State University suggested the use of *high thermal conductivity* carbon foam to distribute the thermal load from the leading edge to the cooler, aft portions of the structure and/or transfer the heat to active cooling features imbedded within the carbon foam (Figure 4). This concept may also take advantage of the preferential thermal conductivity in the z-direction to move the heat aft while partially insulating the interior. AFOSR has expanded upon this concept by placing a layer of thermally conductive carbon foam beneath the CMC shell and insulating carbon foam beneath.

Implementation of carbon foam into these various thermal management systems has been prohibited by its inherent low strength. The material properties reported by the various manufactures of carbon foams are listed in Table 1 and the low compressive strengths clearly stand out. This makes the carbon foam difficult to work with, friable, and subject to physical damage during use. Previous attempts to enhance the strength of carbon foams have focused upon altering the precursors, processing parameters, or upon coating the foam ligaments with a supportive material. Ultramet^{xi}, et al have experimented with coatings of CVD SiC, CVD mullite, CVD Si-B-C, polymers and metals. Each of these approaches has produced composites which have negatively affected the thermal conductivity of the product, added excessively to the mass, or reduced the thermal operational limit. The CVD approach also has a tendency to preferentially deposit on the exterior surfaces of the foam block, leaving a gradient coating, placing a limit upon the cross-section of preforms. Slowing the deposition rate to achieve a more uniform coating, exponentially adds time and cost to the process.

Table 1: Reported carbon foam properties.

PROPERTY	POCO Foam	POCO HTC	MER	Touchstone CFoam
Density (gm/cc)	0.56	0.90	0.19 – 0.80	0.27 – 0.40
Thermal Conductivity (W/mK)				
z-direction	135	245	0.05 – 210	0.25 – 70
x-y direction	78	70		
Compressive Strength (MPa)	2.1	5.9	0.25 – 7.0	4.8 – >15
Total Porosity (%)	70	61	64 – 99	82 – 88
Open Porosity (%)	96	95		
Avg. Pore Diameter (µm)	350	350	30 – 1270	

HEAT EXCHANGER DESIGNS

High thermal conductivity carbon foam can be applied to thermal management systems using two primary approaches. As in most heat exchangers, the carbon foam can become a direct replacement for metal fins in a bypass approach. Here, the foam is bonded to the heat source and fins or pins are machined into the foam to enhance exposed surface area. As shown in Figure 5, the heat transfer coefficient for a metal finned radiator ranges from 30 to 200 W/m²K with a pressure drop of less than 0.05 psi/in. Work by ORNL has demonstrated that a direct replacement of a metal finned heat exchanger with a carbon foam finned heat exchanger can transfer five to thirty times the flux with a near equivalent pressure drop. In applications that call for low pressure drops, such as industrial heat exchangers, vehicle radiators, fan

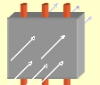
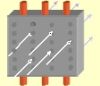
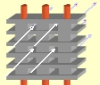
		Heat Transfer Coefficient h, (W/m ² -K)	ΔP/L (psi/in)
Solid Foam		10,000*	2
Through-holes		1,000*	0.1
Finned		1,000*	0.05
Current Radiator		30-200	<0.05

Figure 5: Energy Transfer in heat exchanger configurations.

cooled electronics, and passive convection cooled applications, this approach is imperative. Such systems cannot afford to produce the high pressures (>0.05 psi/in) required for flow-through systems (Figure 6). The latter offer greater volumetric heat transfer efficiency with flux transfer coefficients of up to $10,000 \text{ W/m}^2\text{K}$, but require coolant fluid compression and must afford the costs of the associated equipment in terms of materiel, volume, and mass, and the additional burden of continued operating costs of power and maintenance. These considerations limit the use of the flow-through design and place the selection burden upon the system engineer.

STRENGTH ENHANCEMENT

For either application approach, the friability of the foam challenges the suitability for implementation.

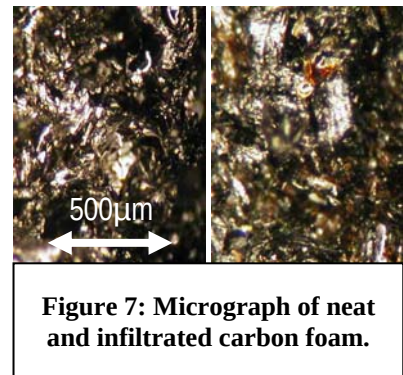
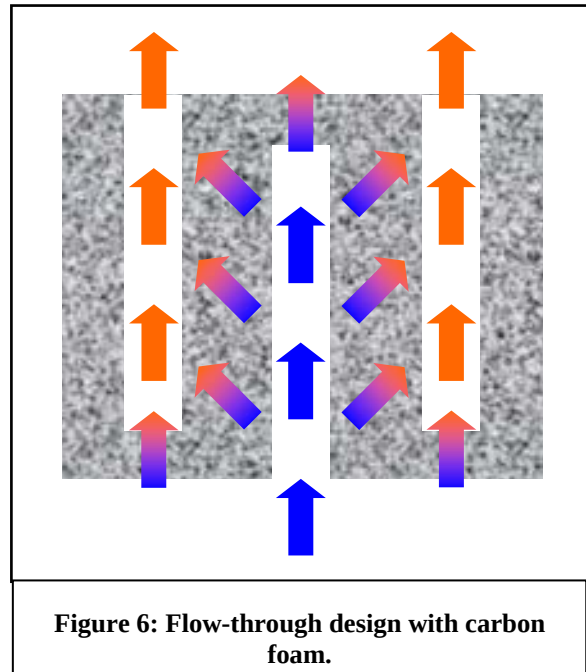
Enhancement Approach

Ceramic Composites Inc. (CCI) elected to take an alternative approach to enhancing the strength of carbon foam. For this evaluation, two polymeric pre-ceramic precursors, which convert to silicon carbide when fired, were selected. Previously used to enhance the fabrication of structural composites materials, these precursors can be fired to a low or high temperature, yielding an amorphous or crystalline beta silicon carbide, respectively. The first polymer is Starfire^{®1}, an allylhydridopolycarbosilane (AHPCS), designated as Polymer A. This polymer modified with the addition of 2% butoxide was also evaluated and designated A+. VL20², a polysilazane designated herein as polymer B was also evaluated. The typical methods of part infiltration is dilution of the precursor with solvent or vacuum infiltration of the preform within a mold, curing at low temperature to release the organic volatiles, followed by optional, repeated infiltrations before firing at an elevated temperature. As the goal was not the formation of a solid second matrix of silicon carbide, but rather a strength enhancement coating, this process was altered to allow the excess infiltrant to drain from the preform prior to curing. Thus, based upon the test parameters listed in Table 2 CCI undertook the study of a wide variety of samples.

Test Results

Micrographs. Microscopic evaluation of the samples revealed a thin coating of silicon carbide on the surface of the carbon ligaments with accumulations of silicon carbide at the ligament fillets as seen in Figure 7b.

Mechanical Testing: The densities of the samples follow the typical trend seen for composite materials where the increase with each subsequent infiltration is reduced (Figure 8). If we were attempting to produce a fully dense sample, the parts would typically be fired to an elevated temperature after the third infiltration before continuing. The difference in density has a dramatic influence upon the ability to flow coolant through the sample and this will be discussed later in this article. Compressive and four-point flexural testing of the samples were performed. Not surprising, the large pore sizes of the samples caused the latter to produce widely scattered data and is not presented. Figure 9 illustrates that the use of a lower concentration of polymer affected the compressive strength, with the 5, 7 and 100% concentrations demonstrating progressive increases. The use of the additive did not appear to affect compressive strength



¹ Starfire Systems, Malta, NY

² Kion Corporation, New York, NY

Table 2: Test matrix variables.

PARAMETER	VARIABLE
Carbon Foam	POCO HTC, POCO Foam
Infiltrates (pre-ceramic polymers)	Polymer A Polymer A plus additive Polymer B
Firing Temperature	Low (amorphous) High (crystalline)
Infiltration Cycles	Zero, One, Two, Three
Infiltrate Concentration	7%, 5% and 100% respectively

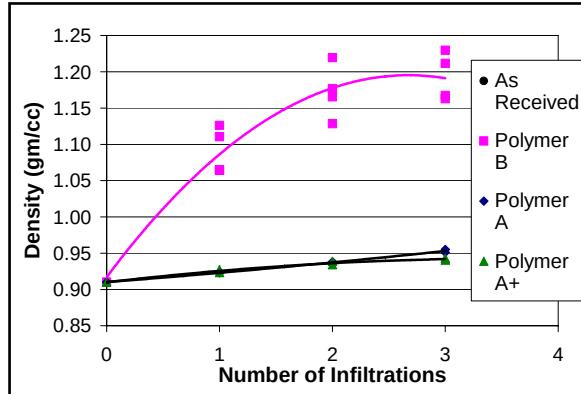


Figure 8: Densities of enhanced POCO HTC Carbon Foam.

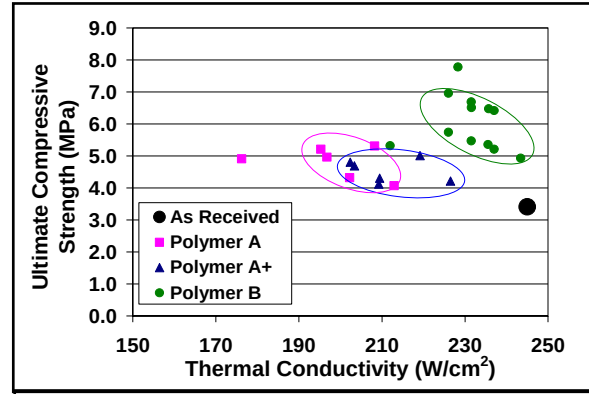


Figure 9: Sample strength and thermal conductivity results.

Hot Plate Testing: The thermal properties of the enhanced carbon foams were measured using the guarded hot plate method. The use of the additive in Polymer A appeared to enhance the bonding between the carbon foam and the ceramic coating, increasing the conductivity despite the lower polymer concentration. Even so, the use of the 100% concentration with Polymer B demonstrated superior performance. Furthermore, firing the B polymer to the higher temperature clearly increased the thermal conductivity of the samples. Little effect of firing temperature was noted for polymer A. However, this could be due to proportionally closer results and a lack of definition in the test method.

Flow Testing: Coolant flow through the carbon foam is of importance to aerospace design engineers as the thermal transfer capability is increased by a factor of ten, compared to flow *past* carbon foam fins.^{xii} Flow testing



Figure 10: Water and gas flow test apparatus.

was performed on 25.4-mm diameter by 25.4-mm thick samples in the x-y direction with both water and nitrogen gas, using the apparatus shown in Figure 10. The data is shown in Figure 11 and reveals a strong relationship between the density of the sample and the ability to flow coolant. The samples infiltrated with Polymer B were unable to flow water, but exhibited an 85% correlation of gas flow rate with density. Select sample infiltrated with Polymer A and A+ exhibited similar or superior flow capabilities to neat foam with both water and gas. The ability to maintain flow through the sample allows the design engineer to perform a trade study between coolant pressurization and heat exchanger performance. It is suspected that the flow-by cooling will be similarly affected.

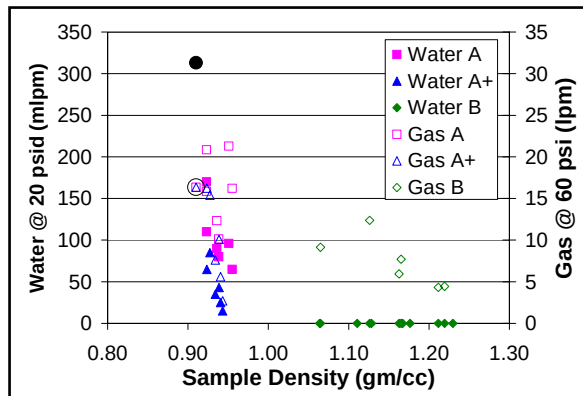


Figure 11: Flow testing data.

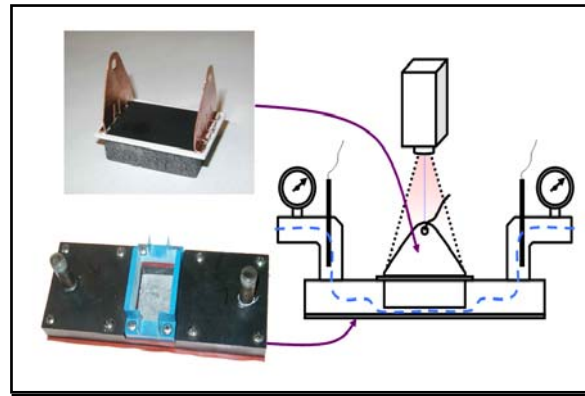


Figure 12: Anteon thermal impedance test fixture.

Thermal Impedance Testing: Anteon Corporation has previously performed testing of the thermal impedance of carbon foam and additional work was performed in this study.^{xiii} Using the apparatus shown in Figure 12, samples of enhanced strength carbon foam which pass coolant in the flow tests were selected for impedance testing. Unfortunately, the Anteon test facility was unable to force coolant water through the samples, so a bypass approach was undertaken. Nevertheless, the results attained were interesting. As water passes below the carbon foam, power is applied to the thick film resistor and the ability of the carbon foam to remove the heat energy was recorded using an infrared camera. This digital data was converted to a thermal impedance value using the formula:

$$\theta_{(x,y)} = \frac{T_{(x,y)} - T_F}{(Q/A_{SAMPLE})} \quad (1)$$

where θ represents the thermal impedance, T the temperature of the sample at point (x,y) or the temperature of the fluid, and (Q/A) , the heat flux. From the resulting data, maps of the heat dissipation of the sample were generated as shown in Figure 11 and thermal impedance plots were developed as a function of water flow rate and applied power as shown in Figure 12. The data for the samples infiltrated with polymer A exhibited a 10 to 33% increase in thermal impedance and, similar to CCI's results for thermal conductivity, the Polymer B samples are projected to exhibit only a 5% increase.

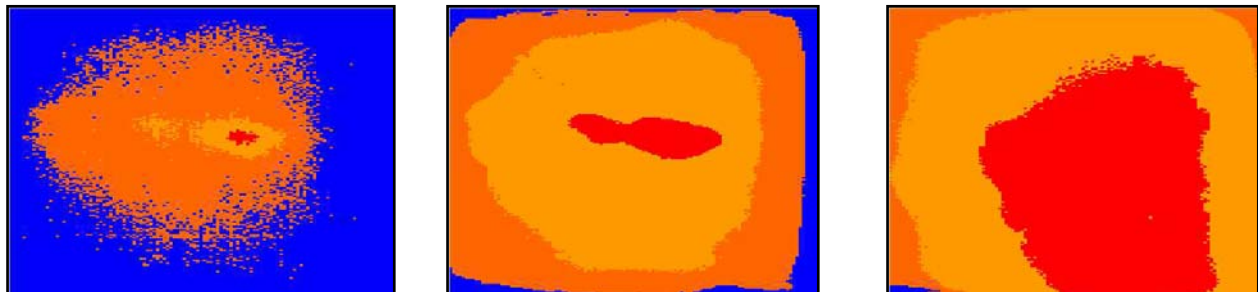


Figure 13: Graphics of thermal impedance testing at 600 W of neat foam at 3.5 and 2.0 gpm and a coated sample at 2.0 gpm.

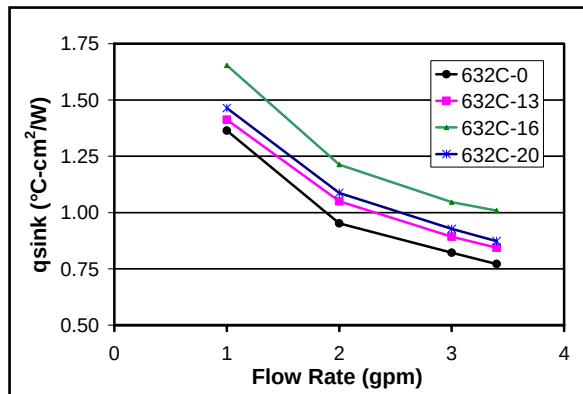


Figure 14: Thermal impedance data.

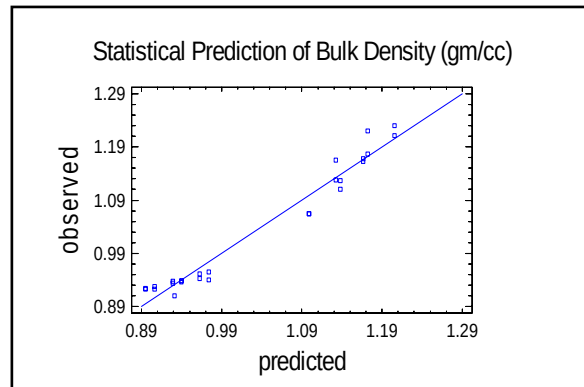


Figure 15: Example of statistical prediction.

STATISTICAL ANALYSIS

Although the quantity of data was insufficient to develop a truly accurate predictive statistical analysis of the relationship between the infiltration processing and the resulting properties, the preliminary analysis presented herein provide insight into the relationships.

Bulk Density

The bulk density of the enhanced POCO Foam is statistically related to the infiltrant, number of infiltrations and first and second order of firing temperature with an R-squared of 96.9% using a backward selection model and an F to enter of 4.0. The accuracy of the predicted and actual values is compared in Figure 13.

$$\begin{aligned} \text{BulkDensity} = & 0.683807 - 0.339437 * \text{StarfireX487} \\ & - 0.331355 * \text{StarfireX487_ZrB} \\ & + 0.0387361 * \text{Infiltrations} \\ & + 0.0000872361 * \text{Temperature} \\ & - 2.45058\text{E-}7 * \text{Temp_sq} \end{aligned} \quad (2)$$

The use of Starfire at 7% or 5% rather than Kion at 100% concentration lowers the bulk density of the structurally enhanced foam by about 0.33 gm/cc.

Compressive Testing

A statistical relationship was not identified for the ultimate compressive strength or the compressive modulus, probably due to high variation in results based upon the large pore volume of the samples. An R-squared of 81% was found for the Strain-to-Failure Ratio using the relationship of:

$$\begin{aligned} \text{Compressive StrainToFailure} = & 0.0225441 \\ & + 0.0137314 * \text{Kion} \\ & - 0.00000843464 * \text{Temperature} \\ & + 0.00435094 * \text{Infil_sq} \end{aligned} \quad (3)$$

Flexural Testing

Between the small number of samples tested and the large variance due to high levels of porosity in the parts, a statistically valid relationship between the flexural strength and the processing was not identified.

Thermal Conductivity

The thermal conductivity is related to the use of Kion as the infiltrant which increased the conductivity by 13 W/m². The relationship between thermal conductivity and the number of infiltrations is a second order function. The net result is a relationship with an R-squared of 82.3%.

$$\begin{aligned} \text{thermalconductivity} = & 117.041 \\ & + 13.1668 * \text{Kion} \\ & - 6.07753 * \text{Infiltrations} \\ & + 5.99142 * \text{Infil_sq} \end{aligned} \quad (4)$$

Water and Gas Flow

Statistical relationships between the processing conditions and the resultant water and gas flows were not identified.

CCI v Anteon Testing

The statistical correlation between CCI's thermal conductivity measurements and Anteon's thermal impedance measurements at 1, 2 and 3 gpm were 81%, 83% and 80%, respectively. However, only six samples are included in the analysis, the correlation, while good, is not sufficiently supported to ensure accuracy. Following continued round-robin evaluation, the more complex Anteon test may be eliminated in favor of the guarded hot plate approach with minimal loss in accuracy.

SUMMARY

Through the application of a silicon carbide coating, via polymeric precursor, to the carbon foam, a uniform ligament coating has been applied to the structure which increases the compressive strength by up to 2.5x and the compressive modulus by up to 3.5x, while reducing the thermal conductivity by only 5%. The comparison is presented graphically in Figure 16. This level of performance enhancement is greater than that known to be achieved by any other means. A tailorable range of properties is achievable with this approach and the approach should be applicable to any commercially available open celled carbon foam. Additional work is suggested to refine the concentrations of polymer and determine their impact upon performance. Finally, with the strength and friability of the foam reduced, it is suggested that effort be shifted from enhancement to application demonstration.

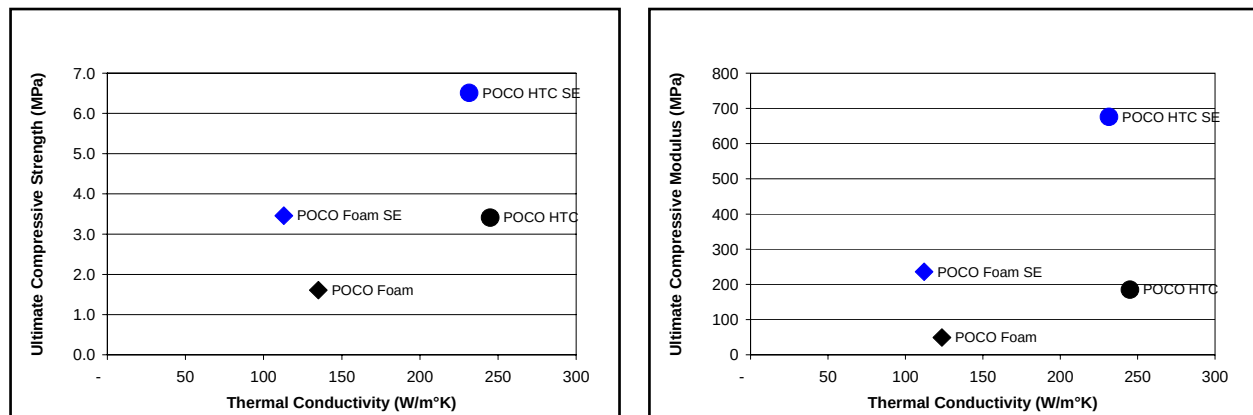


Figure 16: Comparison of the compressive strength and modulus of POCO Foam and POCO HTC with the structurally enhanced modification.

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