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EVALUATION OF ALTERNATIVE FIBERS TO REPLACE NARC-RAYON FOR THE PRODUCTION OF CBCF*

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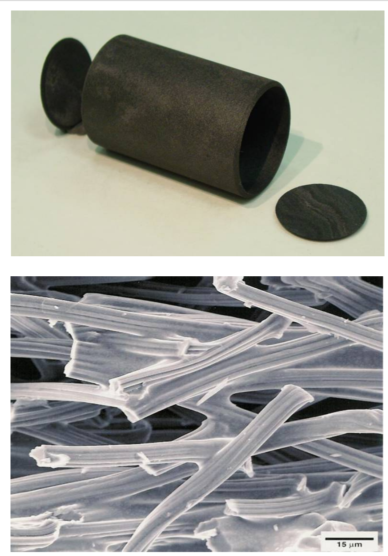
A unique Carbon Bonded Carbon Fiber (CBCF) insulation was developed to provide thermal protection to the isotopic fuel in Radioisotope Power Systems. The microstructure of CBCF is comprised of chopped and carbonized rayon fibers bonded at intersections by carbonized phenolic resin. Production of CBCF insulation at ORNL has been sustained for the past three decades by a single lot of rayon purchased from North American Rayon Corporation (NARC) of Elizabethton, TN in 1987. NARC ceased operations in 1996. Although ORNL has a seven-to-ten-year supply of rayon at the current rate of consumption, an effort has been initiated to identify a new source to meet long-range needs. A summary of findings from the ongoing search for a fiber to replace NARC rayon for production of CBCF is presented.

I. BACKGROUND

The requirement for containment and thermal protection of the isotopic fuel in Radioisotope Power Systems led to a design and material selection for the General-Purpose Heat Source (GPHS) that relied principally on carbon materials. A unique Carbon Bonded Carbon Fiber (CBCF) insulation was developed to provide thermal protection during the extremes of unlikely reentry scenarios. The current configuration of CBCF insulators, shown in Fig. 1**a**, has remained unchanged since its definition for the GPHS[1]. The cylindrical Sleeve and two Discs (endcaps) that make up an insulator set are machined from fully carbonized CBCF preforms. The microstructure of CBCF, shown in Fig. 1**b**, is comprised of chopped and carbonized rayon fibers bonded at intersections by carbonized phenolic resin.

Production of CBCF insulation at ORNL has been sustained for the past three decades by a single lot of aerospace grade rayon purchased from North American Rayon Corporation (NARC) of Elizabethton, TN in 1987. Their principal markets for rayon were for apparel, home furnishings and industrial products. NASA contracted with NARC to produce rayon for solid rocket motor nozzles for the Space Shuttle. Like NASA and other aerospace customers, ORNL stockpiled significant quantities for near- and intermediate-term needs. Due to increased competition from imported rayon, NARC ceased operations in 1996. Although ORNL has a seven- to ten-year supply of rayon at the current rate of consumption, an effort has been initiated to identify a new source to meet long-range needs.



(a) (b)

Fig. 1. (a) CBCF insulator set, and (b) Microstructure of CBCF.

II. CANDIDATE FIBERS

Currently, most of the commercially available rayon is for textile applications or hygiene products. The quality of textile grade rayon fiber is not suitable for CBCF production, either because of chemical impurities or form combined with the fact that very few manufacturers are able to modify their process to meet our specifications. In addition, the relatively low volume requirements (compared to the textile market) for the Radioisotope Power Systems Program makes it an unattractive business case. Despite these challenges, we have identified three candidate fibers that we have started to evaluate.

Fiber-1 is a rayon fiber that is low in sulfur, available in 5.0 single filament denier (versus 1.5 on our current NARC stock), and on a 60-filament tow (versus 160k tow on our NARC stock). We proceeded with evaluation of these fibers despite the higher denier. Fiber-2 is a pure cellulose fiber, produced with a 1.5 single filament denier and 42,300 tow (more similar to our current NARC stock). Fiber-3 is also a rayon fiber, and has become available just recently, so very limited information is available. The single filament denier is 1.7 and is available in various tows.

III. RESULTS

III.A. Thermophysical Properties of Fibers

After chopping the fiber tow to the specified length, we first need to convert the white fiber into a carbon fiber. For that, we must understand the carbonization behavior of the new candidate fibers to ensure the current carbonization profile is still appropriate or if alternative profiles must be evaluated. To this end, a series of differential scanning calorimetry (DSC) and thermogravimetric analyses (TGA) were conducted with the new fiber samples. A summary of the results is presented below.

III.A.1. DSC Studies

DSC analyses of the stock NARC material (Lot J3634) and two of new candidate materials (Fiber-1 and Fiber-2) were carried out at two different heating rates (20ºC/min and 2ºC/min) up to 400ºC which is the instrument limit. The temperature is high enough to observe the various transitions during the pyrolysis process of the fibers. The results are presented in Figs. 2 and 3.

At the faster heating rate (20ºC/min), Fig. 2 clearly shows two endothermic reactions. The first endothermic reaction is likely either the softening point or glass transition point. Fiber-1 and NARC rayon exhibited the transition at a similar temperature, around 106ºC, while the transition for Fiber-2 occurred at a slightly higher temperature, around 122ºC. The second endothermic reaction very likely corresponds to the pyrolysis of the fibers (see results for the TGA analysis), and it occurs at a very similar temperature for all three fibers, ~ 350-360ºC. The reaction is more intense for the NARC rayon, followed by the Fiber-2 and then Fiber-1. The beginning of an exothermic reaction appears to occur prior to the second endothermic reaction, primarily for the Fiber-1 and Fiber-2. If that exothermic peak is confirmed, it may correspond to some type of cross-linking prior to pyrolysis. Additional testing is needed to clarify this.

At the slower heating rate (2ºC/min), which is a heating rate more representative of the heating profile used during the preparation of carbon fibers, the results showed a less-pronounced (almost non-existent) endothermic reaction around 105-122ºC (Fig. 3). This is desirable, since we want to avoid the softening and fusing of fibers during the conversion to carbon fibers, and thus maintaining fiber shape during pyrolysis. The second endothermic reaction, corresponding to the pyrolysis of the fibers, is less pronounced and shifted toward lower temperatures – approximately 320º C (versus 350-360ºC).

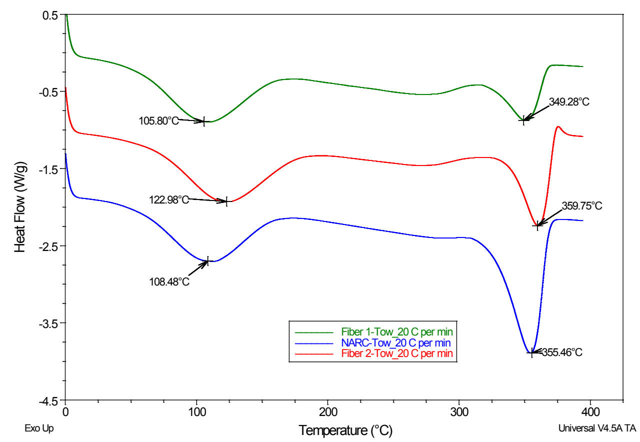


Fig. 2. DSC data at 20°C/min for our NARC rayon and two of the fibers being evaluated for CBCF production.

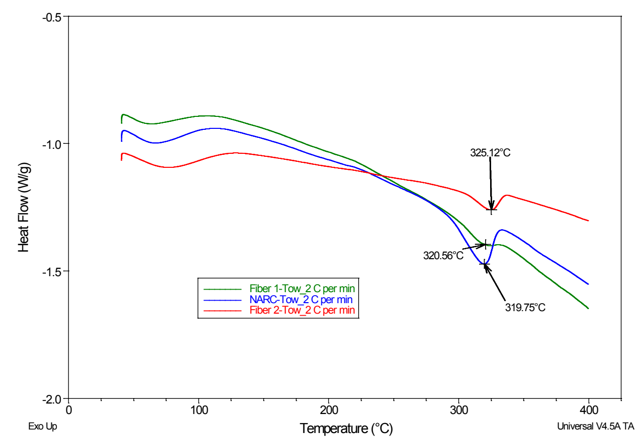


Fig. 3. DSC data at 2°C/min for our NARC rayon and two of the fibers being evaluated for CBCF production.

III.A.1. TGA Studies

TGA tests of the stock NARC material and two of the new candidate materials (Fiber-1 and Fiber-2) were carried out at two different heating rates (20°C/min and 2°C/min), up to the instrument limit of 1000°C. The results for the 20°C/min runs are presented in Fig. 4. The figure shows a peak at the same temperature as the second endothermic peak from the DSC tests. This confirms the peak corresponds to pyrolysis of the fibers and a significant amount of weight is lost during this process. The yield at the end of the endothermic process is about 20%, which is close to our average yield during carbonization (see Table I). As the temperature increases, the yield at the end of the 1000°C segment is closer to 10-15%. This lower yield is probably due to some additional burn-off during the heat treatment caused by the TGA furnace atmosphere not being fully inert due to lower N_2 flow in the TGA system than in the carbonization furnace. At the lower heating rates, the results follow the same trend as those in the DSC, namely the peak for the weight loss is shifted to slightly lower temperatures.

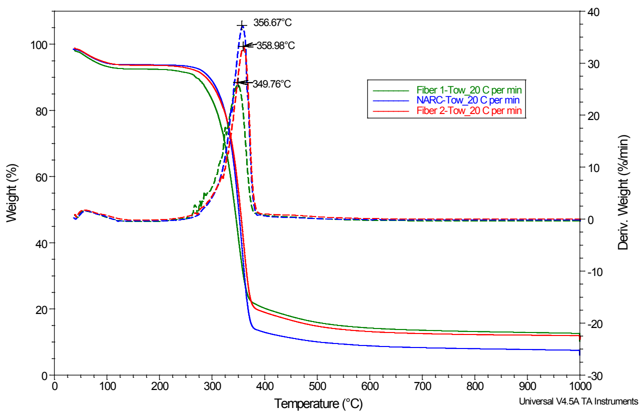


Fig. 4. TGA data at 20°C/min for our NARC rayon and two of the fibers being evaluated for CBCF production.

III.B. Properties of CBCF Produced with Alternative Fibers

Larger quantities of chopped Fiber-1 and Fiber-2 were carbonized in preparation for production of CBCF per our approved procedures. Table I lists the yields during the carbonization of production quantities. Fiber-1 and Fiber-2 exhibited slightly lower yields than our qualified NARC fibers. Two test CBCF Sleeve Lots were produced with carbonized, chopped Fiber-1 (TS-2018-1) and Fiber-2 (TS-2018-3) materials and processed according to our approved procedures for slurry molding, drying, curing, carbonization, sampling, outgassing and testing.

The results for TS-2018-1 (prepared with Fiber-1) are shown in Table II. As shown in this table, the density of the CBCF produced with Fiber-1 is significantly higher than our specification limits, which is reasonable given the higher single filament denier of the fiber. However, all other properties are within the specifications, including the thermal conductivity which normally would have been higher for a CBCF with these high-density values. This may indicate that Fiber-1 materials have much lower thermal conductivity than our current rayon fibers.

The results for TS-2018-3 (prepared with Fiber-2) are shown in Table III. The density values are within the specification limits. Characterization of samples will be completed and presented at the Conference.

TABLE I. Yield after carbonization of the various fibers.

Sample	Yield after carbonization
NARC	23.4
Fiber-1	20.2
Fiber-2	19.8
Fiber-3	*

* Data not available yet.

TABLE II. Properties of CBCF Sleeves produced with Fiber-1 Material.

Property	CBCF Specification[2]	TS-2018-1 Sleeves
Density (g/cc)	0.20 to 0.25	0.294, 0.291
Compressive Strength (psi)	≥ 75 @ 5% strain	105, 101
Ash (wt. %)	≤ 0.3	0.07, 0.18
Sulfur (μg/g)	≤ 300	200, 199
Impurities (μg/g)	Per specification list	All within
Thermal Conductivity (W/m·K)	≤ 0.086	0.0655, 0.0766

TABLE III. Properties of CBCF Sleeves produced with Fiber-2 Material.

Property	CBCF Specification[2]	TS-2018-3 Sleeves
Density (g/cc)	0.20 to 0.25	0.244, 0.242, 0.247, 0.241
Compressive Strength (psi)	≥ 75 @ 5% strain	*
Ash (wt. %)	≤ 0.3	*
Sulfur (μg/g)	≤ 300	*
Impurities (μg/g)	Per specification list	*
Thermal Conductivity (W/m·K)	≤ 0.086	*

* Data not available yet.

II. CONCLUSIONS

Significant progress has been made toward identifying potential alternative fiber sources for production of CBCF. Characterization of the Sleeve lot made with Fiber-2 will continue along with machining of Sleeves for radiographic inspection. CBCF plates will need to be produced, with both the Fiber-1 and Fiber-2 materials to fully evaluate both type of fibers. The new Fiber-3 option adds another potential supplier to this study. These fibers will be processed upon receipt of the test sample.

ACKNOWLEDGMENTS

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