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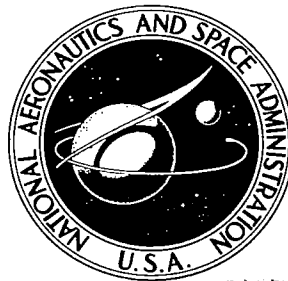
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CARBIDE COATED FIBERS
IN GRAPHITE-ALUMINUM COMPOSITES $(1 \; Feb - 31 \; Jul \; 74)$

Progress Report No. 2:
February 1 - July 31, 1974

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16. Abstract

This report describes the second part of an NASA-supported program at the Los Alamos Scientific Laboratory (LASL) to develop carbon fiber-aluminum matrix composites. Chemical vapor deposition (CVD) has been used to uniformly deposit thin, smooth, continuous coats of TiC on the fibers of graphite tows. Wet chemical coating of fibers, followed by high-temperature treatment, has also been used, but shows little promise as an alternative coating method. Strength measurements on CVD coated fiber tows showed that thin carbide coats can add to fiber strength.

The ability of aluminum alloys to wet TiC was successfully demonstrated using TiC-coated graphite surfaces. Pressure-infiltration of TiC- and ZrC-coated fiber tows with aluminum alloys was only partially successful.

Experiments were performed to evaluate the effectiveness of carbide coats on carbon as barriers to prevent reaction between aluminum alloys and carbon. Initial results indicate that composites of aluminum and carbide-coated graphite are stable for long periods of time at temperatures near the alloy solidus.

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SUMMARY

Chemical vapor deposition (CVD) has been used to uniformly deposit thin, smooth, continuous coats of TiC on the fibers of graphite tows. Wet chemical coating of fibers, followed by high-temperature treatment, has also been used, but shows little promise as an alternative coating method. Strength measurements on CVD coated fiber tows showed that thin carbide coats can add to fiber strength.

The ability of aluminum alloys to wet TiC was successfully demonstrated using TiC-coated graphite surfaces. Pressure-infiltration of TiC- and ZrC-coated fiber tows with aluminum alloys was only partially successful.

Experiments were performed to evaluate the effectiveness of carbide coats on carbon as barriers to prevent reaction between aluminum alloys and carbon. Initial results indicate that composites of aluminum and carbide-coated graphite are stable for long periods of time at temperatures near the alloy solidus.

INTRODUCTION

This report describes the second part of a NASA-supported program at the Los Alamos Scientific Laboratory (LASL) to develop carbon fiber-aluminum matrix composites. The LASL approach to this problem has been to use protective-coupling layers of refractory metal carbides on the graphite fibers prior to their incorporation into the composites.[1] Such layers are directly wettable by liquid aluminum and should act as diffusion barriers to inhibit the formation of aluminum carbide (Al_4C_3). The specific objectives of the work are the following:

● establish the deposition parameters for producing thin, uniform coats of zirconium carbide (ZrC) and titanium carbide (TiC) on carbon, characterize the coat morphology, and determine the extent of damage to the fibers, if any, due to the coating process

● demonstrate the ability of aluminum to wet the carbide surfaces, including the surfaces of carbide-coated graphite, and evaluate the effectiveness of carbide coats on carbon as diffusion barriers to prevent the formation of Al_4C_3

● develop a technique for, and perform liquid Al infiltration on small samples of carbide-coated yarn

● determine tensile strength, elastic modulus and mode of fracture of Al-infiltrated carbide-coated yarns.

Significant progress has been made toward achieving these objectives. During the first part of the program,[1] thin, smooth, continuous ZrC coats were uniformly deposited on individual fibers throughout carbon fiber tows using CVD. Wet chemical methods were also used to produce TiC coats, but with only marginal success. The wettability of ZrC and TiC surfaces with aluminum alloys was successfully demonstrated, and initial attempts to infiltrate ZrC-coated graphite fibers with aluminum looked promising.

In this report the results of the second part of the program are given. Thin, smooth, continuous coats of TiC were uniformly deposited on the individual fibers throughout graphite tows using essentially the same batch CVD process as that used to produce the ZrC coats above. Strength measurements on these materials showed that strength can be increased by the presence of thin TiC coats on carbon fibers. Attempts were made to improve the coats produced by wet chemical methods, but with little improvement over those produced earlier. Further experiments were performed in which the wettability of TiC and ZrC coats on graphite was demonstrated. Numerous attempts have been made to infiltrate carbide-coated fibers with aluminum alloys, using atmospheric pressure, but with little success. However, a pressure-infiltration method has been used, and offers a promising alternative.

EXPERIMENTAL

Materials

Fibers.-The following commercial carbon fibers were obtained from their respective manufacturers for use in coating experiments: Hitron 401 from HITCO, Thornel 50 from Union Carbide Corporation, and HMS from Hercules. Properties and characteristics of these fibers were discussed previously.[1]

Coating Materials.-For the CVD experiments, commercial grade, purified $TiCl_4$ was used. For the wet chemical coatings, tetramethylammonium metalates of Ti, Nb, and Ta were used in aqueous solutions with dextrose. The Ti and Nb solutions contained 0.105 kg of metal and 0.110 kg of dextrose per liter; the Ta solution contained 0.102 kg Ta and 0.116 kg dextrose per liter. (Preparation of the solutions is described in reference 1.)

Aluminum Alloys.-Three aluminum alloys were used for wetting and infiltration experiments. Two of these were the standard commercial alloys, 1100 and 6061, and one was a LASL-produced Al-13 wt % Si alloy. Chemical compositions are listed in Table I.

Fiber Coating Experiments

Two approaches were taken for producing refractory metal carbide coatings on carbon fibers. The first was chemical vapor deposition using a refractory metal halide as the principal constituent of the coating gas. The other was to dip-coat the fibers in a metal-bearing aqueous solution, dry it in air, then convert the coat to carbide by heating at elevated temperatures.

TABLE I

ALUMINUM ALLOYS

Alloy	Composition by Element (wt%)[a]									
	Fe	Si	Cu	Zn	Mg	Mn	Cr	Ti	other	Al
1100	--	1.0[b]	--	0.2	0.1	--	0.05	--	--	0.15max	rem
6061	0.7	0.6	0.25	0.25	1.0	0.15	0.25	0.05	0.15max	rem
LASL Al-Si	0.35	13.4	0.40	0.01	2.45	0.40	--	--	0.18max	rem

[a] Values for 1100 and 6061 alloys are nominal handbook values; those for the Al-Si alloys were determined at LASL.

[b] Combined Fe and Si.

Chemical Vapor Deposition.-$TiCl_4$ was used to produce coats of TiC, on carbon fibers. The vapor coating apparatus, the processing procedures, and the chemistry of the deposition process have been discussed previously.[1,2]

Basically, the following chemical reactions control the decomposition of $TiCl_4$ which leads to the formation of TiC coats on carbon substrates:

$$(1) \quad TiCl_4(g) + 2 H_2(g) + C(s) = TiC(s) + 4 HCl(g)$$

$$(2) \quad TiCl_4(g) + CH_4(g) = TiC(s) + 4 HCl(g).$$
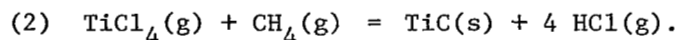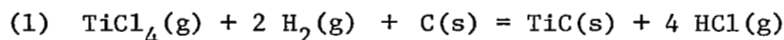
In equation 1, the carbon necessary for the formation of the metal carbide is provided by the substrate which is to be coated. For the reaction in equation 2, the carbon is supplied in the coating gas as methane (CH_4). Besides the coating gases, an inert diluent gas, such as helium is added, together with excess hydrogen. The deposition kinetics can be varied by varying the deposition temperature and the proportions of the gases.

Forty-three CVD runs were made in which TiC coats were deposited on Thornel 50 fibers. In Table II are listed the deposition conditions, the thicknesses and microscopic appearances of the coats, and the tensile breaking loads for the coated fiber tows. The first seven runs (4-8-74 through 4-29-74) were made mainly to coat new carbon fixtures in the CVD apparatus with TiC. Therefore, the carbide coats deposited on fibers included in these runs are not indicative of the coating conditions used. Because of fluctuations in the temperature of the $TiCl_4$ evaporator,[1] it was not always possible to control the delivery rate of the $TiCl_4$ (see Table II). However, as will be shown below, coating gas delivery rate was far less important than the deposition temperature, which was varied from 1023 to 1973 K, and the time which was varied from 5 to 60 min. Both types of deposition reactions were used, as runs were made both with and without CH_4 in the coating gas.

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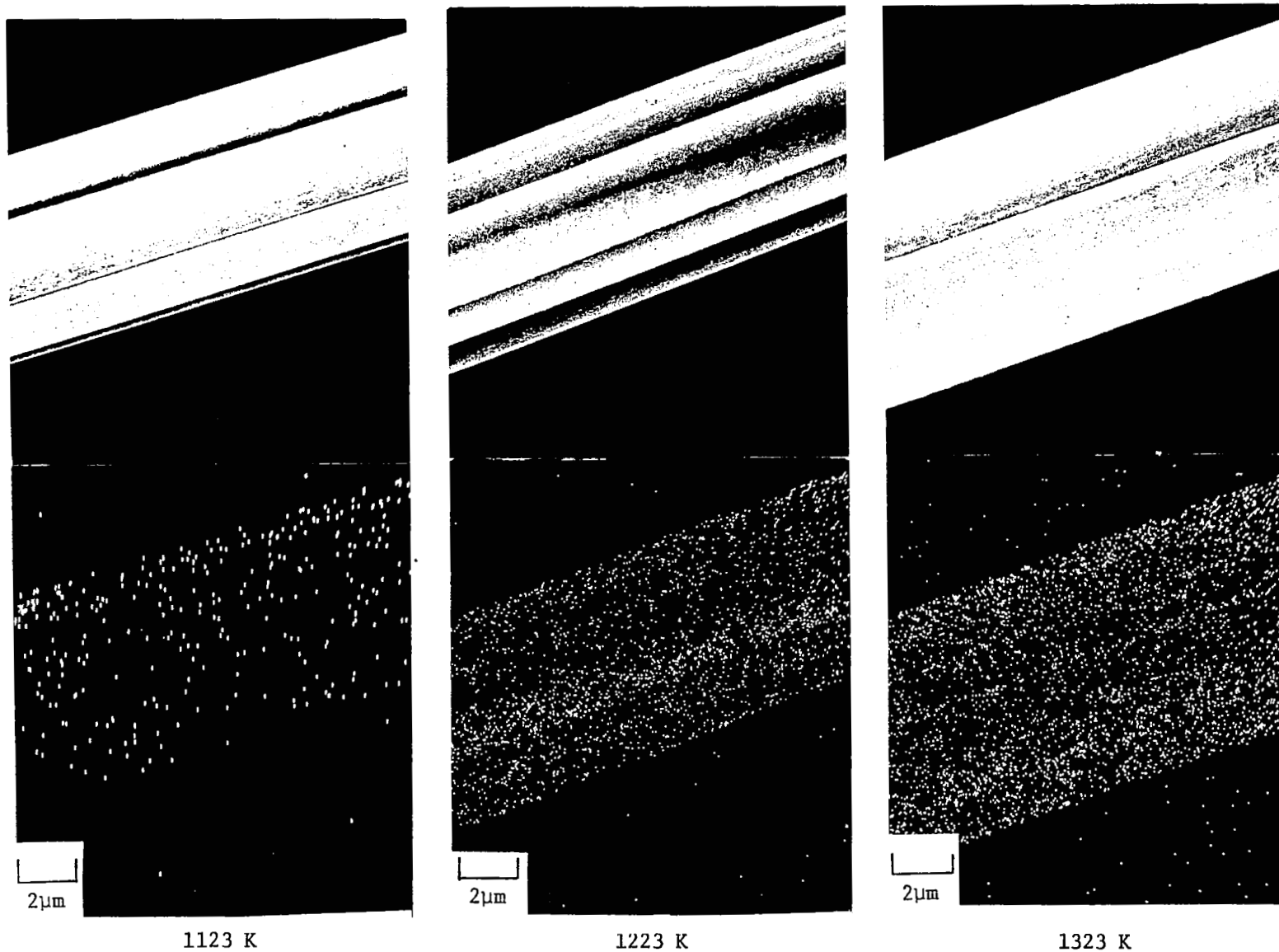1123 K   1223 K   1323 K
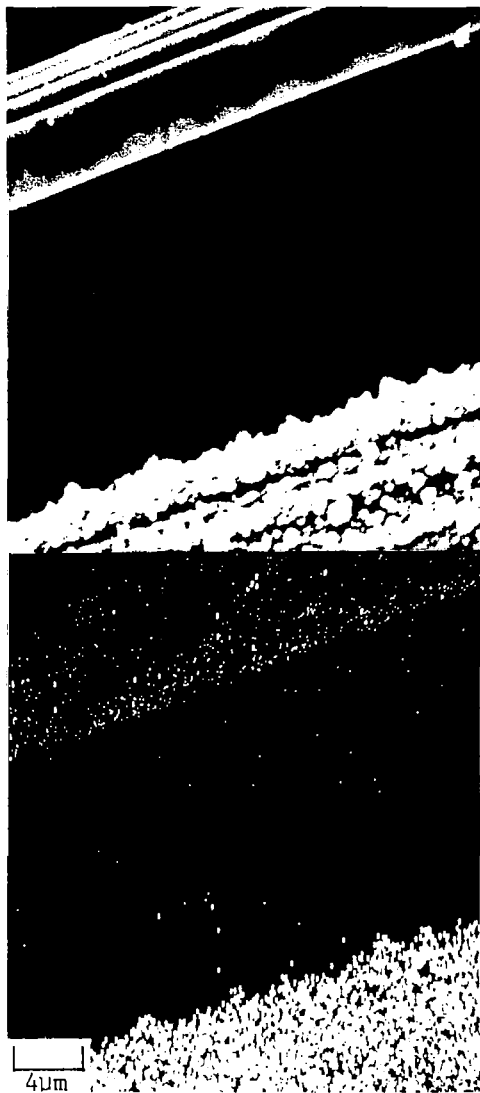
Fig. 1a. SEM speciman images (top row) and Ti x-ray images (bottom row) of Thornel 50 fibers CVD-coated
          with TiC for 60 min at 1123 K (run 6-27-74), 1223 K (run 6-17-74), and 1323 K (run 6-19-74).
          Coating gases contained no CH_4.

1473 K   1723 K   1973 K

Fig. 1b. SEM specimen images (top row) and Ti x-ray images (bottom row) of Thornel 50 fibers CVD-coated with TiC for 60 min at 1473 K (run 5-6-74), 1723 K (run 5-10-74), and 1973 K (run 5-16-74). Coating gases contained no CH_4.

TABLE II

PROCESS CONDITIONS AND RESULTS FOR CARBON FIBER[a] TiC-COATING RUNS

Run No.	Deposition: Temp. (K)	Deposition: Time (min.)	Coating Gas[b] Flow Rate, STP (ℓ/min) TiCl$_4$	CH$_4$	Tow Visual Appearance	TiC Coat max. thk. (µm)	TiC Coat Microscopic Appearance	Tensile Break Load (N) Load	Std. Dev.
4- 8-74	1673	30	c	0	Friable		Nodules, whiskers, mostly unif	7.02[d]	1.86
4- 9-74	1673	30	c	0	Flexible		Few nodules, otherwise unif	4.16[d]	1.28
4-10-74	1673	60	0.09	0	Flexible		TiC occlusions, mostly unif	12.40[d]	3.23
4-15-74	1723	60	0.09	0	Friable		Many nodules to smooth	10.37[d]	1.85
4-17-74	1573	60	0.18	0	Flexible		Few nodules, mostly unif	11.96[d]	4.71
4-18-74	1573	60	0.32	0.15	Flexible		Nodules	10.81[d]	1.51
4-29-74	1573	60	0.25	0	Flexible		Mottled	11.03[d]	1.95
4-30-74	1473	60	0.19	0	Friable			0.79	0.36
5-.1-74	1373	60	0.26	0	Friable	0.3	Uniform, smooth	1.17	0.39
5- 2-74	1173	60	0.27	0	Flexible	0.07	Smooth, non-uniform thru tow	14.78	1.74
5- 3-74	1273	60	0.27	0	Flexible	0.1	Smooth, non-uniform thru tow	10.78	1.41
5- 6-74	1473	60	0.20	0	Friable	0.6	Many nodules, rough	0.46	0.52
5-10-74	1723	60	0.24	0	Friable		Many small crystals, welds	1.02	0.53
5-13-74	1973	60	0.27	0	Friable			1.71	1.03
5-14-74	1023	60	0.23	0	Flexible		Small nodules, many uncoated	11.04	3.10
5-16-74	1973	60	0.31	0	Friable		Large crystals, many welds	0.76	0.49
5-17-74	1673	60	0.24	0.05	Friable		Uniform, many welds	0.41	0.26
5-20-74	1473	60	0.27	0.05	Friable		Uniform, many welds	0.09	0.04
5-21-74	1273	60	0.20	0.05	Flexible		Few nodules to smooth	7.09	2.30
5-22-74	1373	60	0.22	0.05	Flexible		Non-uniform, welds, nodules	2.90	1.03
5-23-74	1273	60	0.21	0.25	Flexible		Nodules, non-uniform	9.12	0.91
5-28-74	1473	15	0.31	0.25	Friable		Few nodules, mostly smooth	1.19	0.77
5-29-74	1973	30	0.30	0	Friable		All converted to TiC	0.10	0.10
5-30-74	1573	60	0.25	0	Friable		Many welds, small crystals	0.19	0.14
5-31-74	1773	60	0.27	0	Friable	1.3	Many welds, small crystals	0.49	0.27
6- 4-74	1273	15	0.24	0	Flexible		Uniform, smooth	11.93	2.29
6- 5-74	1273	60	0.30	0	Flexible		Many nodules to smooth	7.57	1.60
6- 6-74	1273	5	0.21	0	Flexible		Rough to smooth	11.80	1.27
6-11-74	1223	5	0.29	0	Flexible		Smooth, unif, few nodules	10.68	1.26
6-17-74	1223	60	0.33	0	Flexible		Smooth, unif, few nodules	9.87	1.42
6-18-74	1223	15	0.31	0	Flexible		Smooth, unif, few nodules	13.13	1.31
6-19-74	1323	60	0.41	0	Friable		Smooth, unif, few nodules	4.43	1.01
6-20-74	1323	15	0.26	0	Flexible		Smooth, unif, few nodules	10.01	1.34
6-21-74	1323	5	0.38	0	Flexible		Uniform, smooth	12.12	0.80
6-24-74	1273	60	0.36	0.05	Flexible		Uniform, smooth	9.37	1.06
6-25-74	1273	15	0.36	0.05	Flexible		Uniform, smooth	11.71	1.38
6-26-74	1273	5	0.36	0.05	Flexible	0.06	Smooth, unif, few nodules	15.48	1.90
6-27-74	1123	60	0.26	0	Flexible		Uniform, smooth	14.55	2.26
7-10-74	1243	60	0.31	0	Flexible		Mostly small nodules	13.83	1.70
7-15-74	1223	60	0.26	0	Flexible		Few nodules, mostly smooth	11.69	1.16
7-16-74	1273	15	0.26	0	Flexible		Nodules, mottled, smooth	11.13	0.82
7-17-74	1023	60	0.30	0	Flexible			12.10	1.04
7-18-74	1073	60	0.32	0	Flexible			11.85	2.41

[a] Thornel 50 two-ply tow.

[b] Included 5 ℓ/min. H$_2$ and 10 ℓ/min. He in all runs except Run 4-8-74, which had 2 ℓ/min. He.

[c] Unable to determine because of initial equipment set-up problem.

[d] These values not indicative of coating conditions. See text.

In Fig. 1 a typical series of SEM photomicrographs is shown, illustrating the change in TiC coat morphology as a function of deposition temperature. The deposition time was 60 min and the temperature range was 1123 to 1973 K. At temperatures up to about 1373 K there was little change in surface morphology. The coats were thin, smooth and fairly uniformly distributed throughout the tows. Above 1373 K, surface roughness increased rapidly with temperature, and at 1973 K the coat consisted, typically, of large TiC crystals. All the TiC coats shown were produced with a coating gas that did not contain CH_4. However, examination of samples produced with CH_4 showed them to have essentially the same coat morphology for a given coating temperature and time.

In most cases the coats were too thin to be observed by optical microscopy. Therefore, thicknesses were determined on selected samples (Table II) by SEM on cross-sections of epoxy-mounted fiber samples. Figure 2 gives SEM photomicrographs of two typical examples of thin TiC coats prepared in this way.

Figure 3 shows a curve of tensile breaking load vs coating temperature of fibers coated with TiC for a deposition time of 60 min. It includes data for samples whose coats were produced both with and without CH_4 in the coating gas. The testing method was as follows: (1) a small piece of masking tape was folded over a section near each end of the sample of coated fiber tow, (2) the taped ends were clamped between flat parallel grips on an Instron tester, (3) the sample was pulled to failure. At least five samples of each tow were tested.
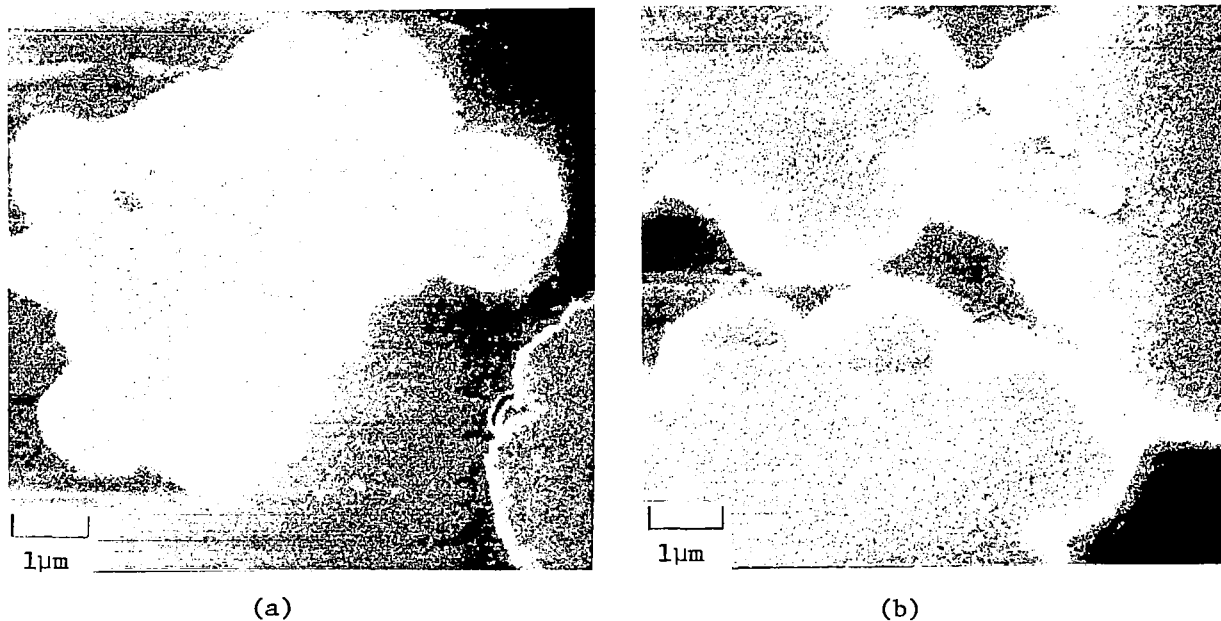

(a)   (b)

Fig. 2. Typical thin TiC coats on Thornel 50 fibers examined by SEM on polished cross-sections of epoxy-mounted samples. (a) Run 7-16-74, (b) Run 7-10-74.
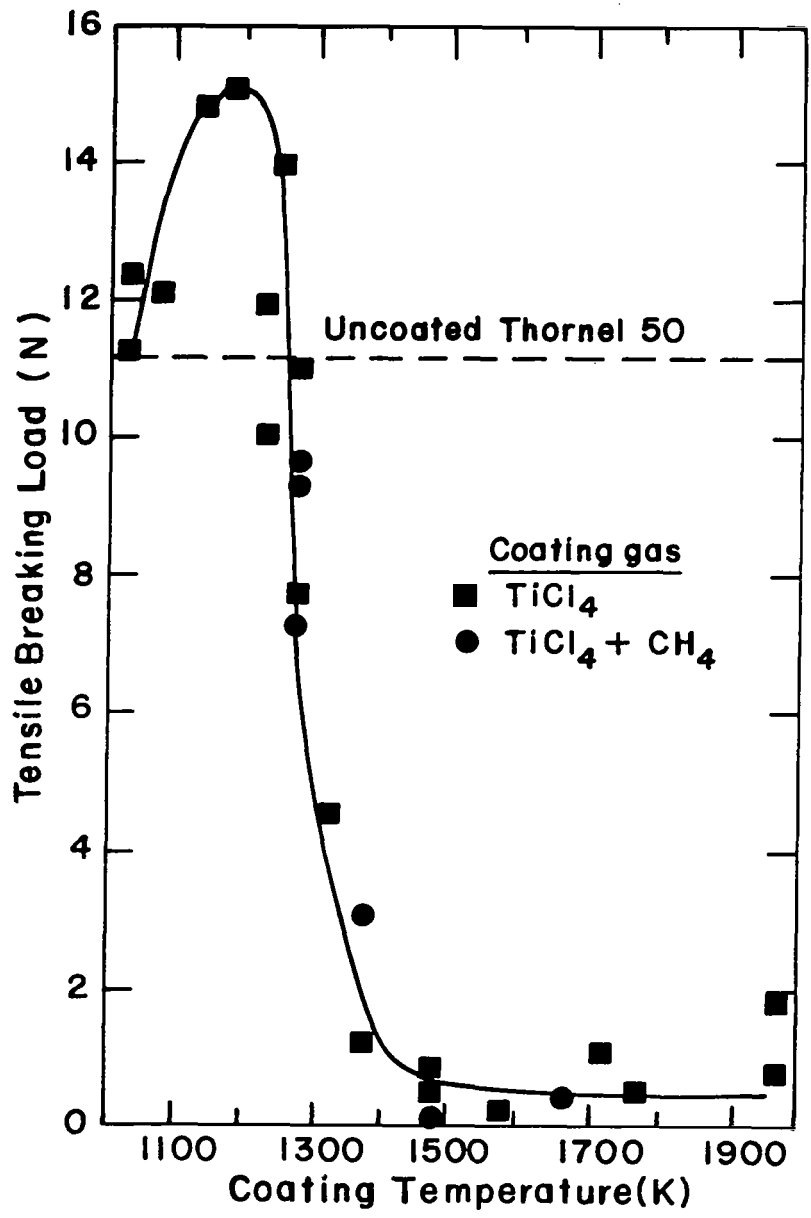
Fig. 3. Tensile breaking load vs deposition temperature for Thornel 50 fibers CVD-coated with TiC. Values are for coats produced both with and without CH_4 in the coating gas. Deposition time was 60 min.

TABLE III

TENSILE BREAKING LOAD OF UNCOATED

THORNEL 50 TOW vs TEMPERATURE

| Temp. (K) | Breaking Load[a] (N) | |
	Mean	Std. dev.
1023	10.34	2.25
1098	12.00	1.51
1173	10.98	1.64
1323	10.66	1.31
1673	12.62	2.05

[a]At least 18 samples of each tow were tested.

To evaluate the effect of temperature, alone, on the strength of the Thornel 50 tow, several uncoated samples were heated at various temperatures in the CVD apparatus without $TiCl_4$ in the coating gas. The breaking loads for these control samples, given in Table III, appear to change little with temperature. Therefore, the average of these values was used as the control for uncoated Thornel 50 and appears in Fig. 3 as a horizontal line.

Figure 3 shows that the strength of TiC-coated fibers increases with increasing temperature up to 1173 K then drops rapidly to nearly insignificant values at about 1473 K. This behavior does not seem to be influenced by the presence of CH_4 in the coating gas, or by the variations in delivery rate of $TiCl_4$ (Table II) during coating. Apparently, the temperature dependence is so great that it tends to mask the effects of variations in these coating process variables. As will be shown below, however, strength is also a function of deposition time.

Figure 3 shows that the strength of carbon fibers can be enhanced by the application of carbide surface coats. However, it has not yet been demonstrated that coats produced under these particular conditions will be thick enough to act as effective diffusion barriers to prevent reaction between carbon and aluminum.

The effect of coating time on the morphology of TiC coats is shown in the SEM photomicrographs of Fig. 4 for samples of Thornel 50 tow coated at 1273 K. This figure includes samples produced both with and without CH_4 in the coating gas. In general, the coats were smooth and uniform, and did not appear to change appreciably within the range of coating times used or with the presence of CH_4.

Figure 5 shows how strength of TiC-coated fibers varied with coating time for three different deposition temperatures. The data given are taken from Table II and include those for samples coated both with and without CH_4 in the coating gas. Some of the points represent the average strengths of samples from as many as five similar coating runs. For the 1223 K coating temperature, strength passes through a maximum at coating time of 15 min. Above 15 min, strength dropped significantly, possibly because of deterioration of the fiber substrate or the development of notch sensitivity in the TiC coat. The curves for the two other coating temperatures, 1273 and 1323 K, may also pass through maxima, but probably at coating times much less than 15 min.

Wet Chemical Coating.—An alternative to CVD coating is a wet chemical coating method.[1] It uses quaternary ammonium metalates which are water soluble and will decompose at red heat to produce metal oxides. When heated at elevated temperature these oxides will convert to carbides if carbon is present. The carbon can

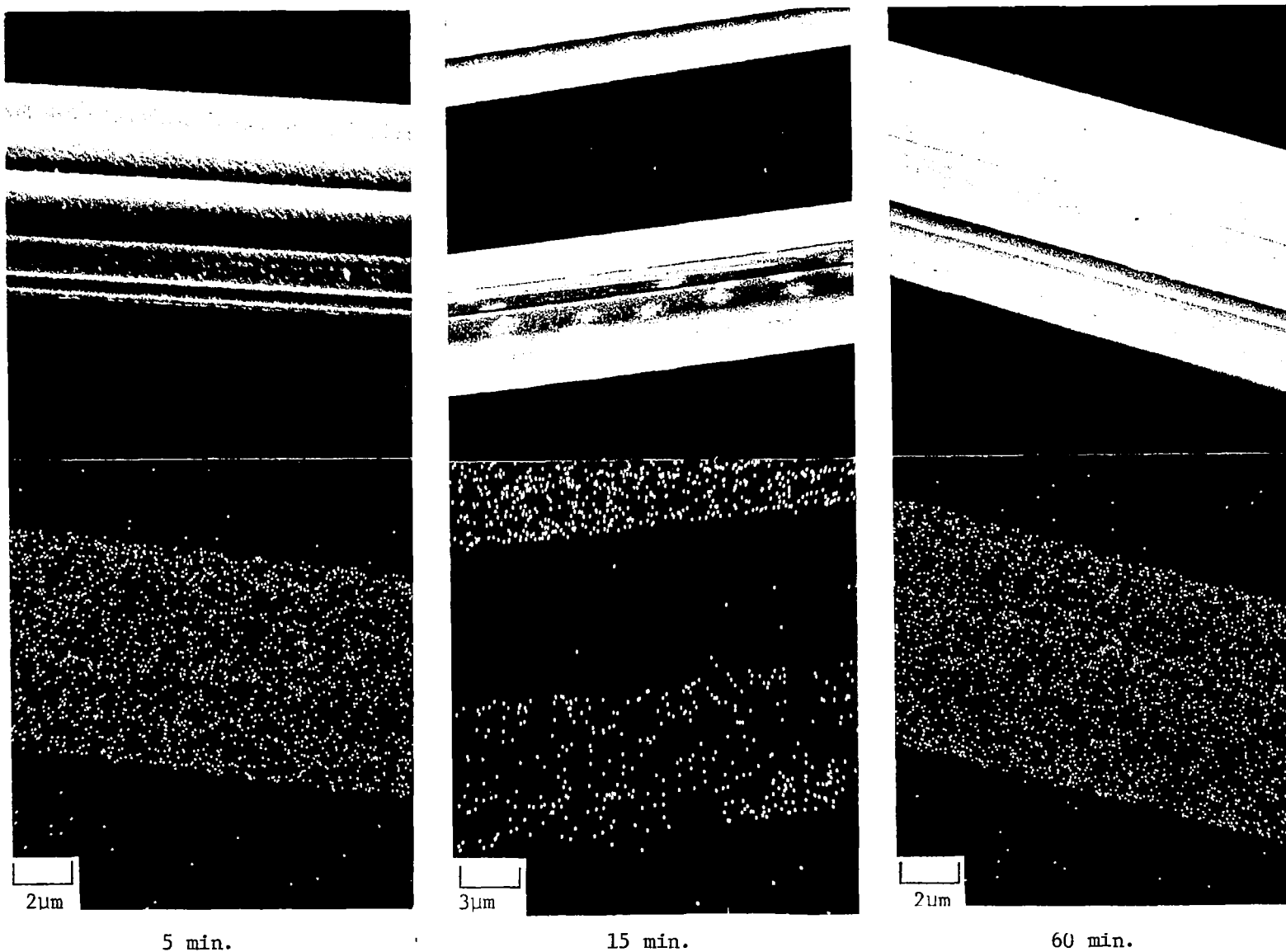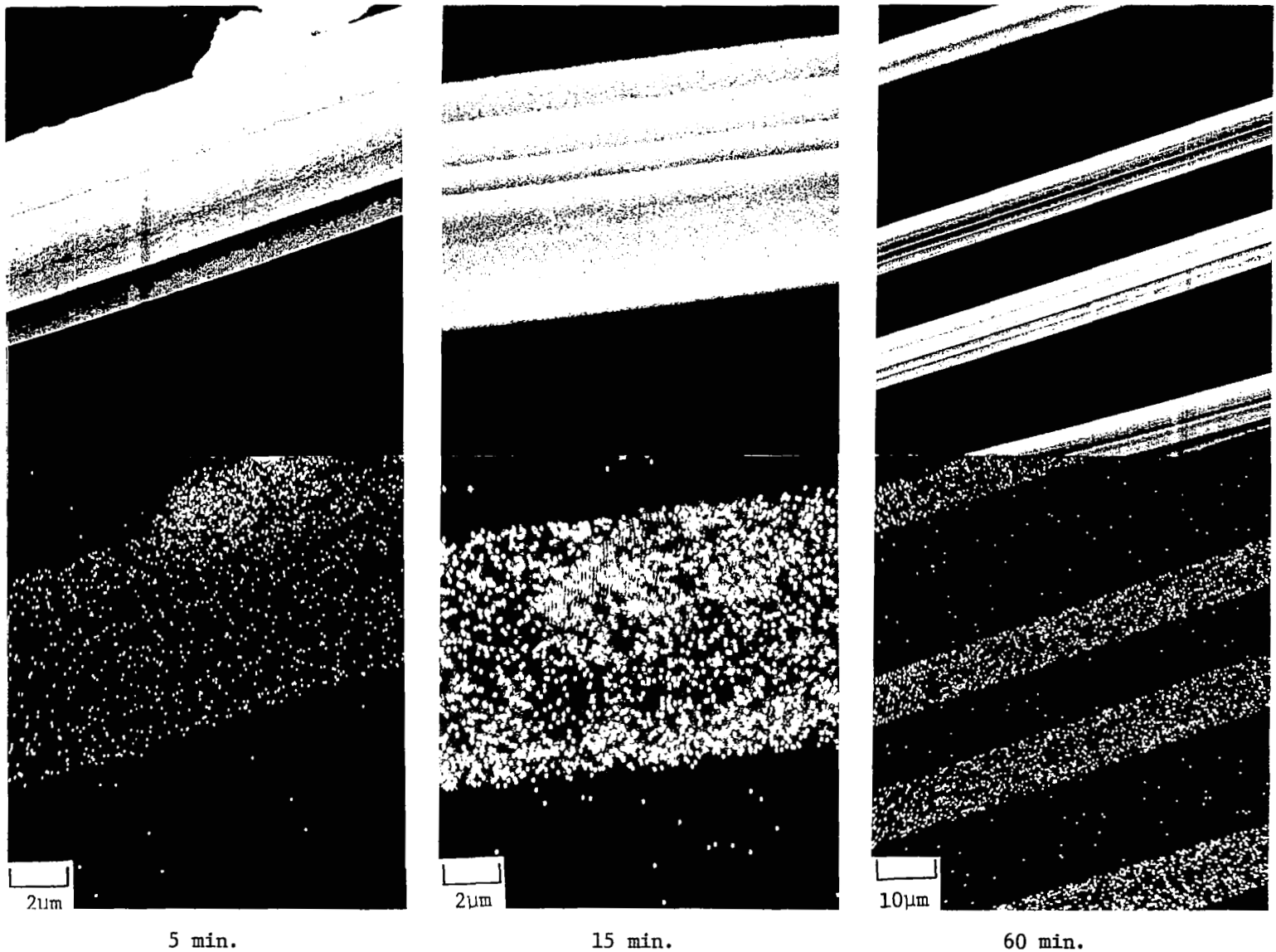5 min.   15 min.   60 min.

Fig. 4a.   SEM specimen images (top row) and Ti x-ray images (bottom row) of Thornel 50 fibers CVD-coated with TiC at 1273 K for 5 min (run 6-6-74), 15 min (run 7-16-74), and 60 min (run 6-5-74). Coating gases contained no CH_4.
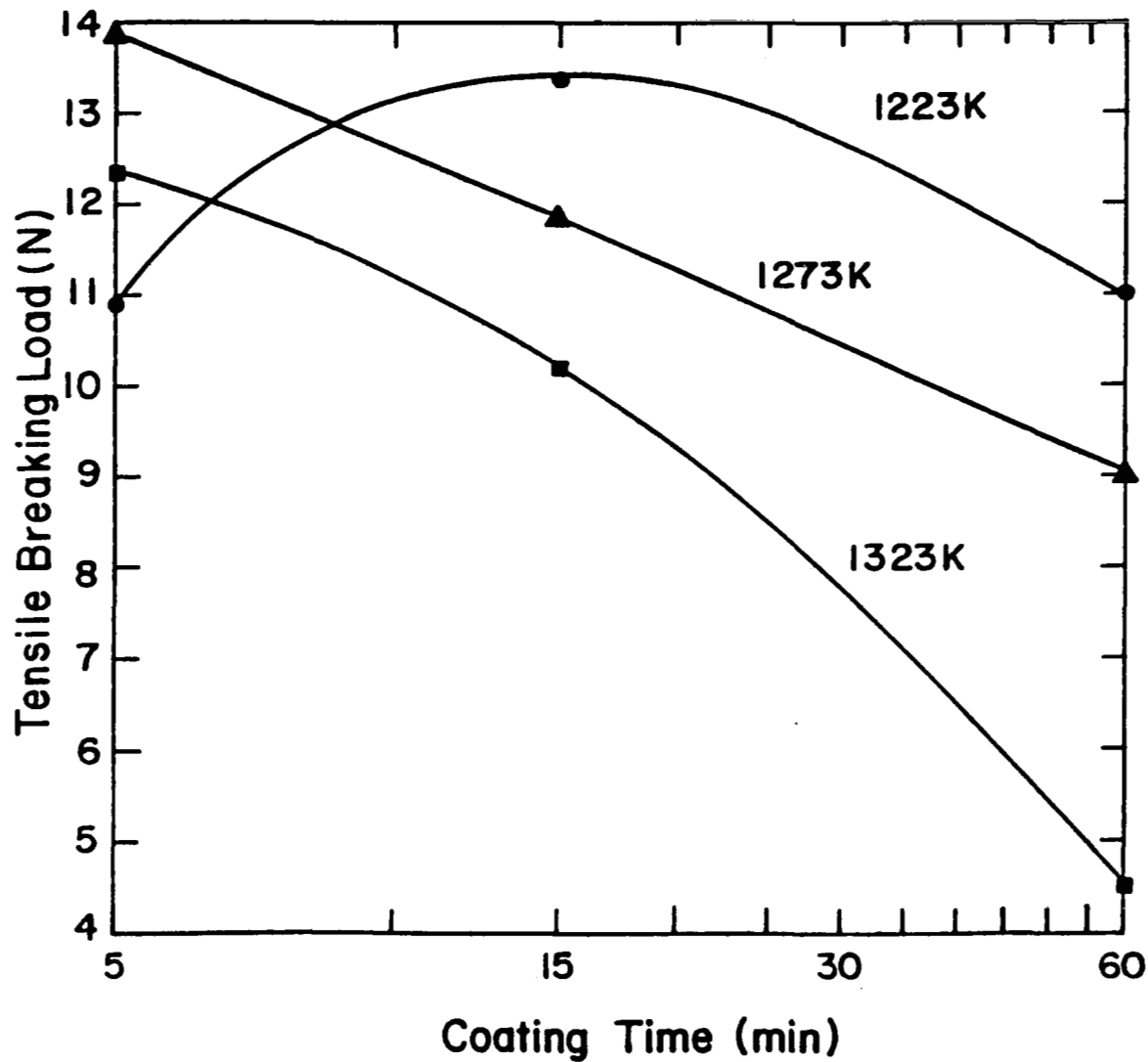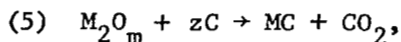
5 min.   15 min.   60 min.

Fig. 4b.   SEM specimen images (top row) and Ti x-ray images (bottom row) of Thornel 50 fibers CVD-coated with TiC at 1273 K for 5 min (run 6-26-74), 15 min (run 6-25-74) and 60 min (run 5-21-74). Coating gases contained 0.05 ℓ/min CH_4.
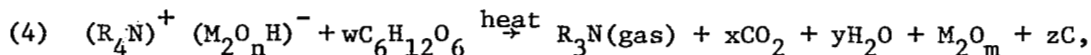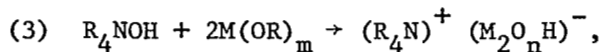
Fig. 5. Tensile breaking load vs time at 1223, 1273 and 1323 K for Thornel 50 fibers CVD-coated with TiC. Values are for coats produced both with and without CH_4 in the coating gas.

be provided by the fiber or by dextrose, which is also water soluble. The chemical equations are roughly:

(3) $R_4NOH + 2M(OR)_m \rightarrow (R_4N)^+ (M_2O_nH)^-$,

(4) $(R_4N)^+ (M_2O_nH)^- + wC_6H_{12}O_6 \xrightarrow{heat} R_3N(gas) + xCO_2 + yH_2O + M_2O_m + zC$,

(5) $M_2O_m + zC \rightarrow MC + CO_2$,

where R is an organic radical and M is a metal. Such metalates are ill-defined, however, and the amount of dextrose needed to supply all the carbon must be determined experimentally. Excess carbon will result in a carbide dispersed in a carbon matrix while too little carbon will cause the metal to convert fiber carbon to carbide. Despite these difficulties, encouraging results were achieved earlier[1] with this system. Therefore, further attempts were made to produce coats by wet chemical methods.

Samples of HMS carbon fibers were dip-coated in aqueous solutions of dextrose containing Ti, Nb or Ta. Each sample was given three dip cycles as follows: (1) dipped in the solution, (2) finger squeegeed between two layers of thin latex rubber, (3) dried at 398 K for 15 min in air. After the three dip cycles, the samples were heated to 1973 K in helium. The only difference between this procedure and that used previously[1] is the finger-squeegeeing step. The typical appearance of fibers coated in this way is illustrated in the SEM photomicrographs of Fig. 6. These coats are just slightly better than those produced previously by wet coating methods,[1] but are still not as uniformly distributed as are those produced by chemical vapor deposition.

Thermal Cycling of ZrC-Coated Fibers.-Because of the difference in the coefficients of thermal expansion between a fiber and its carbide coat, damage to the coat or the fiber surface may occur during thermal cycling. To evaluate this possibility a series of thermal cycling experiments was made in which samples of Thornel 50 and HMS fibers, ZrC-coated in run 12-7, described previously,[1] were heated from room temperature to 1023 K at 10^{-3} Pa. The heat-up time was approximately 11 min and cool-down took 65 min. For up to five cycles, no damage occurred to the coats on any of the fibers examined. Figure 7 shows SEM photomicrographs of typical samples of the ZrC-coated HMS fibers before any thermal cycling (Fig. 7a) and after five cycles (Fig. 7b).

<center>WETTING EXPERIMENTS</center>

Previously, ZrC surfaces were successfully wetted with aluminum alloys using the fixture shown in Fig. 8.[1] The fixture was made from graphite and is used as follows. With the sample and alloy located as shown, the fixture is placed in a vertically-oriented, quartz-tube, vacuum furnace. The furnace is heated rapidly to a temperature above the melting point of the alloy, at a pressure of 1×10^{-3} Pa. The plunger is then pushed into the extruder causing a stream of fresh, molten metal to be directed onto the carbide surface. Several additional runs
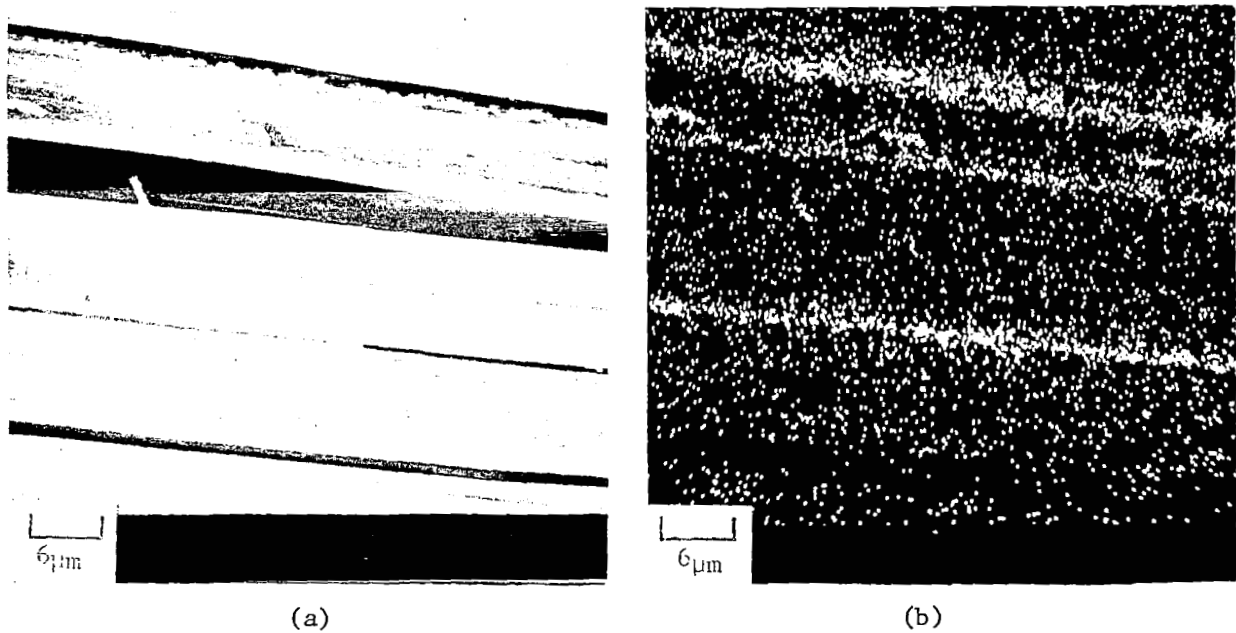
(a)   (b)

Fig. 6.   SEM photomicrographs of HMS fibers wet-chemical-dipped three times in Nb solution, dried between each dip at 398 K, and finally heated to 1973 K.   (a) Specimen image, and (b) Nb x-ray image.

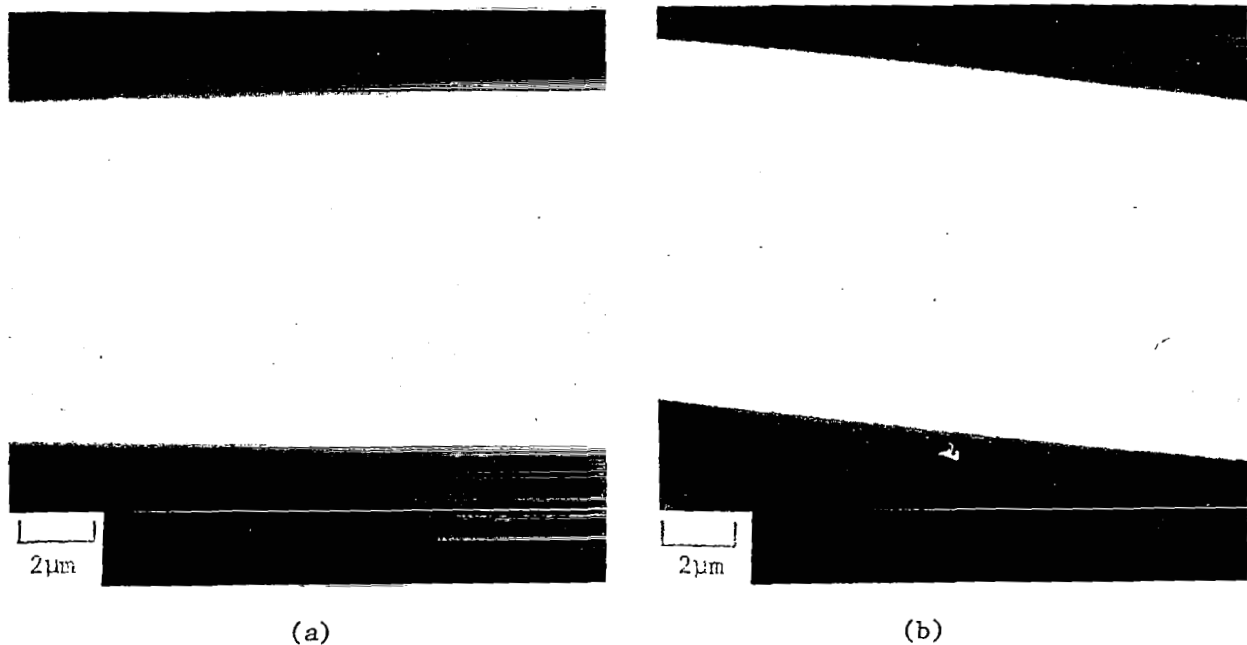
(a)   (b)

Fig. 7.   SEM photomicrographs of HMS fiber CVD-coated with ZrC showing the absence of coat damage resulting from thermal cycling between 293 and 1023 K.   (a) No thermal cycling, and (b) Five cycles.
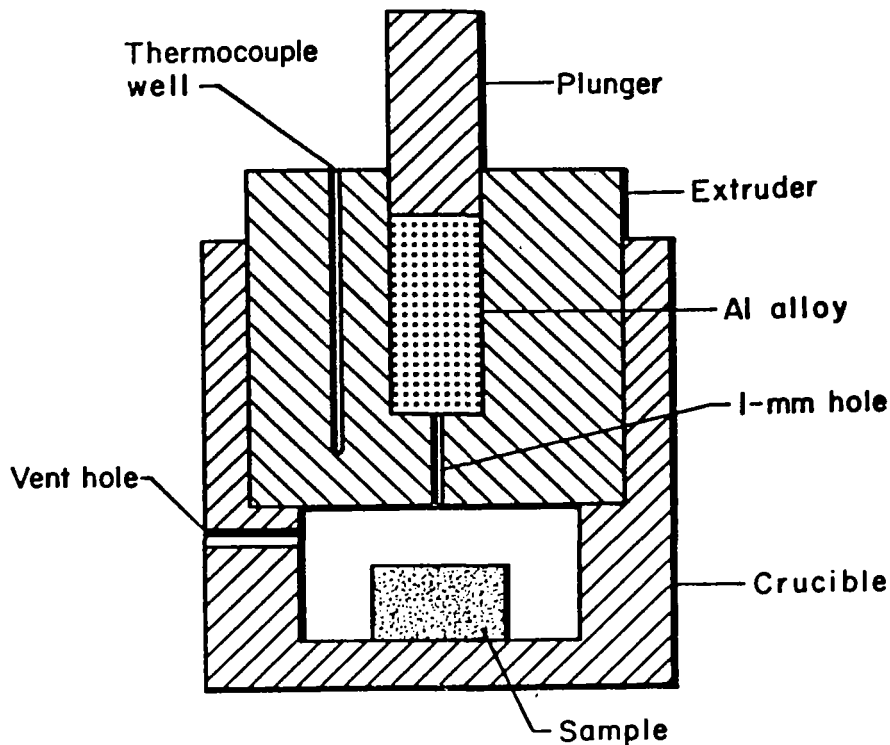
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Fig. 8.   Schematic of fixture for wetting refractory metal surfaces with fresh
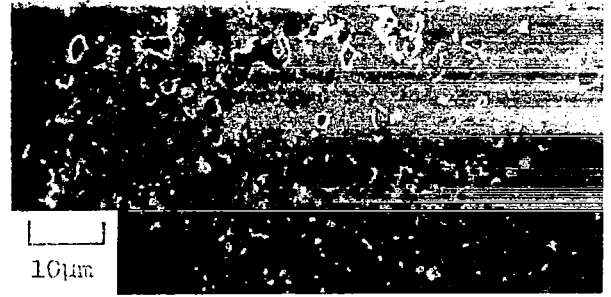   molten aluminum alloys.

were made with this fixture, in which samples of ZrC- and TiC-coated POCO HPD-1
graphite were wetted with the three aluminum alloys listed in Table I. Micro-
scopic examination of the alloy-carbide interfaces of these samples showed good
wetting. A photomicrograph of a ZrC-coated graphite sample wetted with Al-13% Si
alloy is shown in Fig. 9a.

DIFFUSION BARRIER EXPERIMENTS

     To evaluate the effectiveness of a ZrC coat as a diffusion barrier to pre-
vent reaction between aluminum and carbon, the Al-wetted,ZrC-coated graphite
samples, described above in "WETTING EXPERIMENTS," were heated in vacuum (10^{-3} Pa)
under various conditions of time and temperature. In Table IV the heat-treatment
conditions are summarized together with the diffusion couple compositions. For
comparison, samples of uncoated Thornel 50 fiber tow, infiltrated with Al-13% Si
by Fiber Materials, Incorporated, were included in two of the experiments. In
experiment AD-1 (Table IV), the heat-treatment temperature was held at approx-
imately 45 K below the alloy solidus for 100 h, and no reaction appeared to
occur at the alloy-ZrC interface, as shown in Fig. 9b. For experiment AD-3,
similar materials were held at approximately 105 K above the solidus of the

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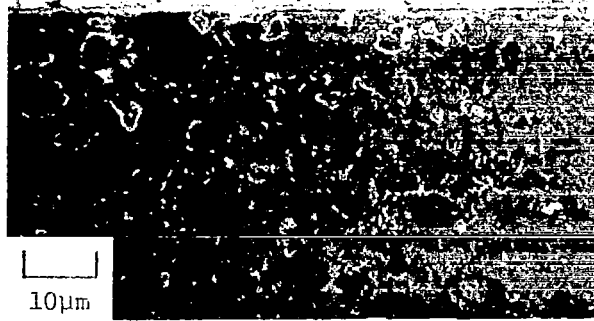
Fig. 9. Optical photomicrographs of typical ZrC-coated graphites wetted with aluminum alloys, illustrating the effect of heat-treatment on the Al/ZrC interface. (a) No heat-treatment, (b) 100 hours at 45 K below the alloy solidus (848 K), (c) 112 hours at 105 K above the alloy solidus (848 K), (d) 150 hours at 13 K below the alloy solidus (916 K).

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TABLE IV

DIFFUSION BARRIER EXPERIMENTS

Exp't	Diffusion Couple (Alloy/Substrate)	Alloy Wetting Temp. (K)	Alloy Solidus Temp. (K)	Time (h)	Heat Treatment Temp (K)	Results
AD-1	Al-13%Si/ZrC-coated graphite	963	843-848[a]	100	803	No Reaction
	Al-13%/Thornel 50 (FMI)[b]	---	---	100	803	No Reaction
AD-2	1100 Al/ZrC-coated graphite	1008	916[c]	150	903	Reaction
AD-3	Al-13%Si/ZrC-coated graphite	963	843-848[a]	112	953	Reaction
	1100 Al/ZrC-coated graphite	1008	916[c]	112	953	Reaction
	6061 Al/ZrC-coated graphite	998	855[c]	112	953	Reaction
	Al-13%Si/Thornel 50 (FMI)[b]	---	---	112	953	Reaction

[a] Determined by thermal arrest on heating during infiltration experiments.

[b] Uncoated Thornel 50 fiber infiltrated with Al-13%Si alloy by Fiber Materials, Incorporated.

[c] From Metals Handbook, 8th Edition, Vol. 1 (1961).

Al-13% Si alloy, and extensive reaction occurred at the interface. This is shown in Fig. 9c by the presence of a dark phase together with some blocky material. As shown in Fig. 9d, it was not necessary to exceed the solidus temperature to produce reaction. Here, the ZrC-coated graphite, wetted with 1100 Al alloy, was held 150 h at 13 K below the solidus. Reaction obviously took place, as evidenced by the presence of a dark speckled material at the alloy-carbide interface.

INFILTRATION EXPERIMENTS

Nearly fifty unsuccessful attempts were made to infiltrate carbide-coated fibers with aluminum using a pressure of 75.8 kPa (1 atm) and a method described previously.[1] Figure 10 gives an example of a typical attempt to infiltrate ZrC-coated Thornel 50 with 6061 aluminum alloy at this pressure. There was essentially no penetration of the yarn tow, although wetting on the outer fibers occurred.

To overcome this problem, an apparatus for pressure-infiltration was constructed, based on a design described by Banker,[3] and shown schematically in Fig. 11. The apparatus is designed so that the casting and melting crucibles can be pressurized or evacuated independently. The infiltration procedure is as follows. The apparatus is placed in a vertically-oriented, resistance-heated, tube furnace. The melting and casting crucibles are both evacuated to a vacuum of about 1 Pa, and the infiltrant alloy is melted. The bottom of the melting crucible is punctured with the spear and the alloy flows into the casting mold containing the carbon fibers. The apparatus is pressurized with argon to pressures
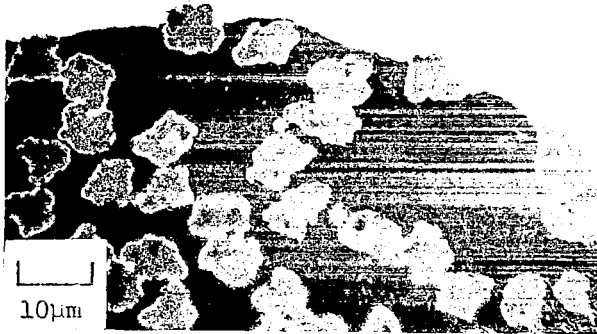
Fig. 10. Optical photomicrograph
showing lack of penetration
of ZrC-coated Thornel 50 tow
by aluminum alloy at 75.8 MPa.

up to 0.35 MPa, held for up to 15 min, then allowed to cool under pressure to room temperature. The mold material is removed from the infiltrated samples by machining.

Several pressure-infiltrations have been made with this apparatus and have shown some improvement over those done at one atmosphere. But, as shown in Fig. 12, penetration of the aluminum alloy into the carbide-coated fiber tow is still poor. However, wherever the alloy comes into contact with the carbide coat the wetting is good. For uncoated fibers, essentially no wetting occurs (Fig. 12b).

CONCLUSIONS

1. Chemical vapor deposition was used to produce smooth, uniform coats of TiC on individual fibers throughout graphite tows. Strength measurements on these materials showed that strength can be increased by the presence of thin coats on the fibers.

2. Surfaces of TiC-coated graphite were successfully wetted with aluminum alloys.

3. Experiments were performed to evaluate the effectiveness of carbide coats on graphite as barriers to prevent reaction between aluminum alloys and carbon. Initial results indicate that composites of aluminum and carbide-coated graphite are stable for long periods of time at temperatures up to 803 K, or about 40 K below the Al-13% Si alloy solidus.

4. Infiltration of TiC- and ZrC-coated fiber tows with aluminum alloys was attempted at pressures up to 0.34 MPa. At low pressures, only the outer fibers of a coated tow were wetted with alloy. At the higher pressures some penetration of the tow occurred, but was still far from complete.
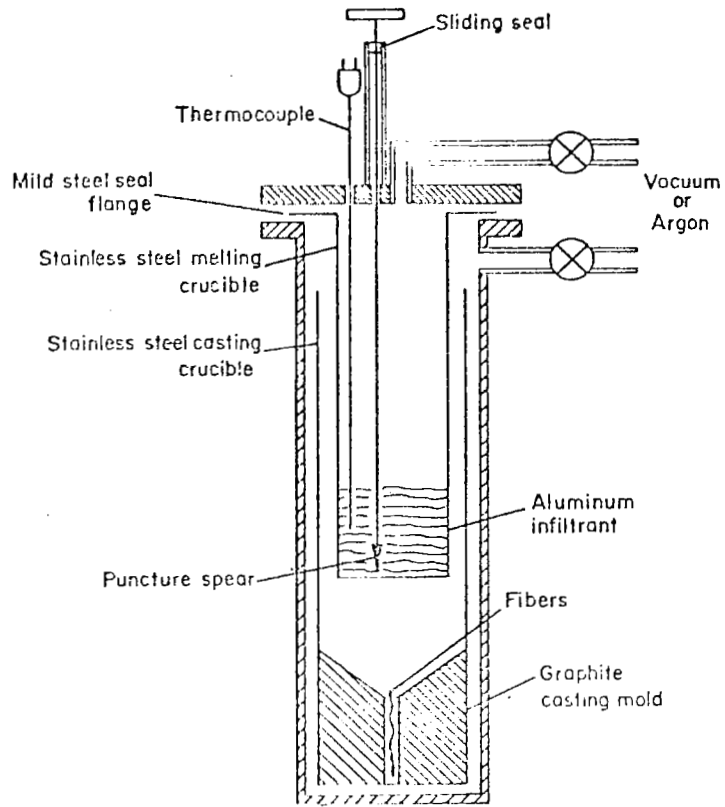
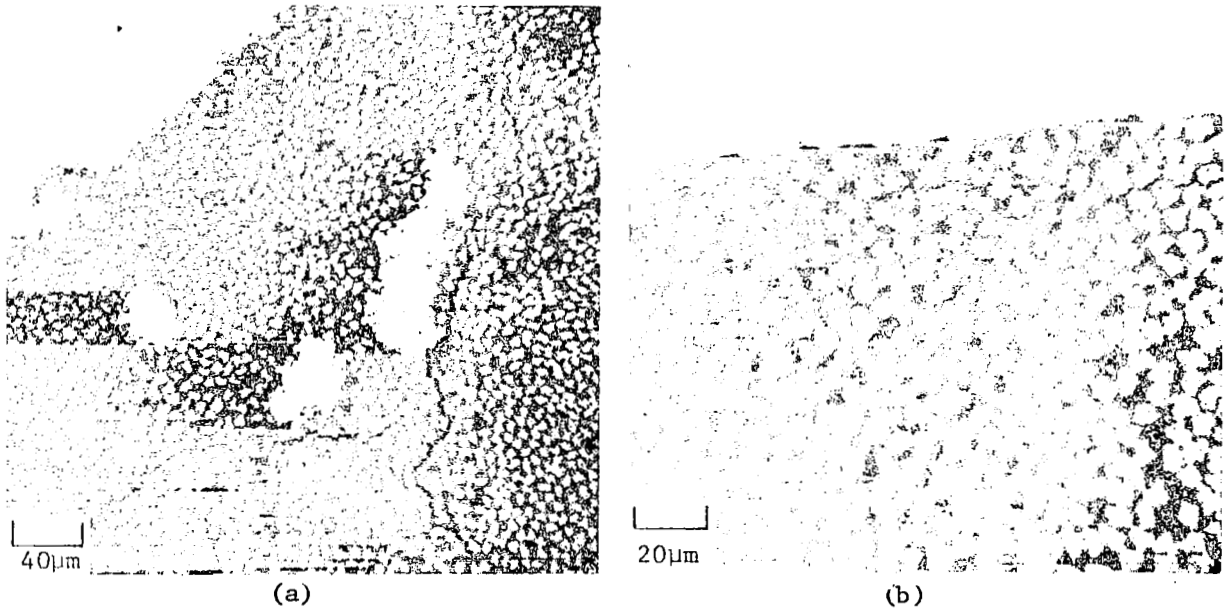Fig. 11.   Schematic of pressure-infiltration apparatus.

40μm   20μm

(a)   (b)

Fig. 12.   Optical photomicrographs of samples of Thornel 50 tow run in pres-
          sure-infiltration apparatus with Al-13% Si alloy at 0.34 MPa and
          948 K   (a) TiC-coated fibers.   (b) Uncoated fibers.

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3. Banker, J. G., "Metal Matrix Composite Fabrication by Liquid Infiltration," SAMPE Quarterly, January 1974, p. 39.