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REVIEW OF THERMAL PROPERTIES OF

GRAPHITE COMPOSITE MATERIALS

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ABSTRACT

Flammability, thermal, and selected mechanical properties of composites fabricated with epoxy and other thermally stable resin matrices are described. Properties which were measured included limiting-oxygen index, smoke evolution, thermal degradation products, total-heat release, heat-release rates, mass loss, flame spread, ignition resistance, thermogravimetric analysis, and selected mechanical properties. This paper describes the properties of eight different graphite composite panels fabricated using four different resin matrices and two types of graphite reinforcement. The resin matrices included: XU71775/H795, a blend of vinylpolystyrylpyridine and bismaleimide; H795, a bismaleimide; Cycom 6162, a phenolic; and PSP 6022M, a polystyrylpyridine. The graphite fiber used was AS-4 in the form of either tape or fabric. The properties of these composites were compared with epoxy composites. It was determined that the blend of

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vinylpolystyrylpyridine and bismaleimide (XU71775/H795) with the graphite tape was the optimum design giving the lowest heat release rate.

INTRODUCTION

Graphite-reinforced composites have potential applications in advanced aircraft and aerospace vehicles because of their weight saving and performance characteristics. Performance characteristics of composites are dependent on the properties of the materials comprising the composite and the process by which they are combined. This rule is particularly true of graphite-reinforced composites, when the thermal and flammability properties are dependent on the type, amount, and orientation of the fiber, and the type and amount of the resin matrix used.

The purpose of this paper is to review the thermal and selected mechanical properties of composites fabricated with epoxy and other thermally stable resin matrices. Properties which are reviewed include limiting-oxygen index, smoke evolution, thermal degradation products, total-heat release, heat-release rates, mass loss, flame spread, ignition resistance, thermogravimetric analysis, and selected mechanical properties. This paper reviews the properties of different graphite composite panels fabricated using different resin matrices and types of graphite reinforcement. The resin matrices reviewed include: epoxy; blends of vinylpolystyrylpyridine and bismaleimide; bismaleimide; phenolic; and polystyrylpyridine resins. The graphite fiber used was a high modulus graphite in the form of either tape or fabric. The properties of these composites are compared with epoxy composites. This review indicates that thermally stable resins such as blends of vinylpolystyrylpyridine and bismaleimide provide the lowest heat release rates which is an important parameter in the design of composite structures for aircraft and aerospace vehicles.

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RESIN CHEMISTRY AND COMPOSITE FABRICATION

Four types of resin matrices were evaluated: a) XU71775/H795

(Hercules, Inc.), a bismaleimide/vinylpolystyrylpyridine (VPSP) formulation;

b) H795 (Technochemie GMBH), a bismaleimide; c) Cycom 6162 (Cyanamid Co.), a

phenolic; and d) PSP 6022 (Societe Nationale et Poudres Explosifs), a poly-

styrylpyridine. Graphite composites made from these resin matrices were

compared with a composite made with an epoxy resin as a matrix.

1. Epoxy Resin: The baseline epoxy resin was an amine-cured polyfunc-

tional glycidyl amine-type epoxy resin. The chemistry of this and the other

resins is shown in Fig. 1.

2. XU71775/H795: This formulation is based on a formulation of bis-

maleimide (H795) and a modified VPSP designated as XU71775.01L (Dow Chemical

Co.). This resin has the same oligomer backbone (polystyrylpyridine) as

VPSP, but possesses different reactive end groups. The chemistry of the

VPSP is shown in Fig. 1 and has been described previously (1,2,3). The

XU71775/H795 formulation contains seven parts by weight H795 and three parts

by weight XU71775.01L. Other reactive materials are added to this formula-

tion to allow hot melt prepregging of the resin. This resin was charac-

terized thermally by differential scanning calorimetry (DSC), thermogravi-

metric analysis (TGA), and exothermicity. The DSC of the resin was measured

at 10°C/min in nitrogen and is shown in Fig. 2. The endothermic peak at

60-140°C is probably due to the evaporation of trace amounts of volatiles.

The exothermic cure temperature is at 160-240°C with a cure peak at 211°C.

The resin can be cured at 177°C for a longer time. After heating at 177°C

for 3 hr, the resin showed no residual cure peak. The exothermic peak at

300-360°C is probably related to the decomposition of the resin. The TGA of

the cured pure resin in nitrogen at 10°C/min is shown in Fig. 3. The resin

starts to decompose at about 320°C. The char yield at 800°C is 52.5%. The

viscosity of the resin was determined by obtaining gel curves in the Rheom-etrics apparatus. Viscosities were 800,000 poise at 25°C, 1170 poise at 50°C, 6.2 poise at 100°C, and 2.7 poise (minimum) at 127°C. The gel times for the resin were 5 min at 177°C, 18 min 40 sec at 140°C and greater than 1 hr 15 min at 121°C. The resin exothermed in 10 hr at 80°C and 3 hr at 100°C. The resin is soluble in tetrahydrofuran and dimethylformamide, and is dispersible in methyl ethyl ketone.

3. H795: This bismaleimide resin is produced by reacting m-maleimidobenzoic acid chloride with an aromatic diaminocompound in the molar proportion of difunctional amine acid halide 1.4:2. The resulting resin consists of a mixture of a bismaleimide and an aminoterminated monoimide as shown in Fig. 1. This mixture, close to the eutectic mixture, is cured by melting at 120° to 140°C which causes polymerization by addition of the free-amino groups to maleimide double bonds followed by a vinyl poly-merization of the terminating maleimide double bonds. The H795 is a brit-tle, glassy solid which becomes tacky and flexible above 50°C. To obtain a room temperature drapeable and tacky prepreg, the resin requires formulation with a reactive diluent to avoid losing mechanical properties at elevated temperatures. Thus, H795 was mixed with a reactive, unsaturate, high-boiling liquid monomer. This mixture was soluble to at least 70% by weight in methyl ethyl ketone. A free-radical inhibitor was added to reduce the reactivity of this formulation. The resulting formulation possessed physi-cal and reactivity properties which were suitable for hot-melt and solvent-based prepregging. The DSC of this resin formulation shows that a small exotherm (4 J/g) occurs at 105-155°C, with a major exotherm (200-250° J/g) at 214°C.

The resin exotherm started at about 10 hr at 100°C. The TGA shows about 5% weight loss in volatiles up to 160°C, with major decomposition

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occurring at 415°C. Char yield is 55% at 520°C. The viscosity at 78°C is 1200 poise with minimum viscosity of 4.2 poise at 130°C. The gel time at 177°C is 8 to 9 min. The resin is soluble in acetone, methyl ethyl ketone, methylene chloride and is insoluble in 1,1,1-trichloroethane.

4. Cycom 6162: The exact chemistry of this commercial phenolic resin is not known. A probable structure is shown in Fig. 1.

5. PSP 6022: The reaction scheme for this resin is shown in Fig. 1. This resin has been described previously in detail (4).

The resins just mentioned were used to fabricate eight types of composite panels using two types of reinforcements: a) plain-weave woven graphite fabric (A-193, Hercules Inc.), and b) unidirectional tape graphite fiber (AS-4, Hercules Inc.). All panels were fabricated using a honeycomb core (HR-10, Hexcel Inc.). All composites had a film of polyetheretherketone (PEEK, Imperial Chemical Co.) adhered with a silicone adhesive (Dow Corning X3-5815) on one side.

The composition of the panels is given in Figs. 1 and 4. The thickness of the panels varied slightly depending on the number of plies in each panel. Panels constructed with the graphite fabric had 1 ply on each side and panels fabricated with the graphite tape had 3 plies on each side placed at 0°, 90°, and 0° orientation for maximum strength. The processing of the baseline panel consisting of epoxy-glass fabric with a polyvinylfluoride (PVF) film has been described previously in detail (5). Panels type A, B, E, and F were cocured with the honeycomb core without the use of an additional adhesive. Panels type C, D, G, and H used a polyimide adhesive film (FM-34, Cyanamid Co.) to bond the face sheets to the honeycomb.

The XU71775/H795 prepreg used for panels type A and B is prepared by roll milling the resin onto the graphite fabric. To prepare composites, the

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prepreg is precured 20 min at 130°C, cured 6 hr at 177°C, and postcured 18 hr at 177°C.

The H795 fabric prepreg for panel type C is prepared using a solvent coater with the resin dissolved at 65% by weight in acetone. The tape for panel type D is prepared by hot-melting the resin. To prepare composites the prepreg is cured 2 hr at 177°C and postcured 1 hr at 204°C and 4 hr at 232°C. The prepregs for panels type E, F, G, and H are available commercially and their curing schedules have been reported previously in Masaline (1979) and in Cycom 6162 (Technical Bulletin, Cyanamid Co.).

The resin content from the aforementioned prepregs was determined by extracting it with dimethylacetamide. The resin content, R, was determined by

$$R = [(w_p - w_f)/w_p] \times 100 - V_o \qquad (1)$$

whereby w_p = weight of prepreg, w_f = weight of fibers, and V_o = volatile content. The resin content of the composites fabricated using the above prepregs was in the range of 38-42% by weight.

The curing schedule for panels type C, D, G, and H is given in Table 1. The composite laminates were precured prior to bonding to the honeycomb core with the polyimide film adhesive. The curing schedule for panels type A, B, E, and F is given in Table 2. These panels were cocured with the honeycomb core without the use of an additional adhesive. The panel's adhesiveness is attributed to the inherent adhesive properties of the prepregs used in these composite panels. The fabrication procedure for all the composites has been reported previously in detail (6).

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THERMAL AND FLAMMABILITY PROPERTIES

A broad range of flammability and thermal tests were conducted to characterize the composites. Six basic thermal and flammability properties of the materials were measured: 1) propensity to burn, or oxygen index (OI), 2) smoke emission, 3) heat release, 4) toxic-gas emission, 5) ignition resistance, and 6) surface flammability. In some cases, more than one test apparatus was used to measure the same property, thus allowing a comparison of test methods. The thermal stability of the composites was determined by TGA and by exposure to a radiant heat source to determine mass loss.

The composite panels were tested by the oxygen index in accordance with ASTM D-2863-77 (7). The intent of the oxygen index test method is to determine the relative flammability of plastics by measuring the minimum concentration of oxygen in a slowly rising mixture of oxygen and nitrogen that will just support combustion; i.e., oxygen index is defined as the minimum concentration of oxygen and nitrogen that will just support combustion of a material under conditions of this method. The test results are given in Table 3. Panel D-H795 Tape/PEEK had the highest oxygen index of all the panels tested followed by Panel A-XU71775/H795 Fabric/PEEK. The baseline-epoxy glass fabric/PVF had the lowest oxygen index.

The smoke emission characteristics of the panels were determined using two different methods. The first method involves heating the composites in the National Bureau of Standards (NBS) chamber using the technique of smoke accumulation (8). The second method involves heating the composites in the Ohio State University (OSU) heat-release apparatus (9). The first method subjects 76.2- × 72.2-mm specimens mounted in a vertical steel holder to a radiant energy source of 2.5 W/cm^2 for a period up to 20 min. The smoke evolved is measured through a light path of 91 cm in the sealed chamber using a light source and photomultiplier tube arrangement.

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The percentage change in the light transmission is converted to an optical density value by means of the following equations:

$$D_s = (V_1/AL)\log_{10}(100/Plt) \qquad (2)$$

where

D_s = specific optical density

V_1 = chamber volume, 0.51 m^3

L = light-path length, 0.91 m

A = exposed-test-specimen surface area, 42.35 cm^2

Plt = percent light transmission

The smaller the D_s value, the better the material. The smoke-test data are included in Figs. 5 and 6 and give D_s of the composites at specific times during the test. Composite panel type G (PSP 6022/Fabric/PEEK) had the lowest smoke evolution at 1.5 min and the baseline epoxy-glass fabric/ PVF had the highest smoke evolution at 1.5 min. Panel type H was not tested. In the second method, from OSU, the specimen to be tested is injected into an environmental chamber through which a constant flow of air passes. The specimen's exposure is determined by a radiant heat source adjusted to produce the desired total heat flux on the specimen of 3.5 W/cm^2. The specimen is tested so that the exposed surface is vertical. Combustion is initiated by a piloted ignition. The smoke is measured with a photoelectric tube mounted on top of the apparatus. The smoke density is calculated by integrating the light transmission loss over the length of the run.

$$\text{Specific optical density, max} = D_s = \frac{V_2}{AL} \int_0^t \log_{10} \frac{(100)}{(Plt)} \frac{T_o}{T_i} \, dt \qquad (3)$$

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where

t = time

V_2 = volume of air, 2.4 m^3/min

A = area of sample, 232.3 cm^2

L = length of light path, 0.93 m

Plt = percent of light transmission

T_i = inlet temperature

T_o = outlet temperature

The test results are given in Fig. 7. As in the previous tests, the baseline epoxy-glass fabric/PVF composite had the highest smoke evolution of all the composites tested. Panel type H (PSP 6022 Tape/PEEK) had the lowest smoke evolution. When comparing the two test methods, the relative ranking of the composites in terms of increased smoke density at 90 sec is as follows: NBS smoke method, panels type D, G, E, F, A, C, B, and Baseline; OSU smoke method, panels type H, B, D, F, C, A, E, G, and Baseline. The two test methods do not correlate very well.

The heat release of the composite panels was determined using the OSU Release Calorimeter (9) using a revised test method (10). In this procedure, the specimen to be tested is injected into the environmental chamber through which a constant flow of air passes. The specimen's exposure is determined by a radiant heat source adjusted to produce the desired total-heat flux on the specimen of 3.5 W/cm^2 using a calibrated calorimeter. The temperature difference between the air entering the environmental chamber and that leaving is monitored by a thermopile having three hot and three cold, 32-gauge Chromel-Alumel junctions. The hot junctions are spaced across the top of the exhaust stack. The cold junctions are located in the pan below the lower air-distribution plate. Heat-release rates are

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calculated from the reading of the thermopile output voltage at any instant of time as

$$HRR = \frac{(V_m - V_b) \times K_h}{0.02323 \ m^2} \qquad (4)$$

where

HRR = heat-release rate, kW/m^2

V_m = measured thermopile voltage, mV

V_b = "blank" thermopile voltage test obtained by a run conducted with an empty sample holder assembly

K_h = calibration factor, kW/mV

The integral of the heat-release rate is the total-heat release. According to regulations for aircraft (11) the total-heat release over the first 2 min of sample exposure shall not exceed 65 $kW \cdot min/m^2$, and the peak-heat-release rate shall not exceed 65 kW/m^2. Figures 8 and 9 give the total-heat release and peak-heat release rate of the composite panels when exposed at a heat flux of 3.5 W/cm^2. According to these test results, of the nine panels tested only panel type B (XU71775/H795 Tape/PEEK) met the earlier criteria. The total-heat release of this composite was 62 $kW \cdot min/m^2$ and the peak-heat release rate was 51 kW/m^2. Panels type A and H with heat release of 66 and 67 kW/m^2, respectively, were marginal failures.

The samples were tested for carbon monoxide (CO), nitrogen oxide (NO_x), hydrogen fluoride (HF), and hydrogen cyanide (HCN). These gases are measured using calorimetric detector tubes as approximate parts per million (ppm), produced during the flaming mode in the NBS smoke chamber described previously. The test results are given in Table 4. The baseline panel showed the highest CO evolution while NO_x, HF, and HCN was approximately the same in all the composite panels. Type H panel was not tested. The limits

of these gases fall within the guidelines established by the aircraft industry.

The composite panels were tested for resistance to ignition using the procedure described previously (12). The composite panels were evaluated for after-flame time and burn length. The specimens were conditioned in accordance with the Standard. The test criteria for this test are as follows: burn-length maximum average, 15.2 cm; after-flame maximum average, 15 sec; and drip-burn maximum average, 3 sec. The results are given in Table 5. The panels, except panel type A (XU71775/H795 Fabric/PEEK), when tested with respect to drip burn, after flame time and burn length, passed this test. Panel A failed because the burn length extended beyond 15.2 cm.

The surface flammability of the composites was determined using a radiant energy source. This method of measuring surface flammability of materials essentially employs a radiant heat source and an inclined specimen. The orientation of the specimen is such that ignition is forced near its upper edge and the flame front progresses downward. The incident heat flux to the specimen ranged from a maximum of 4.4 W/cm^2 at the top to a minimum of 0.4 W/cm^2 at the bottom. A factor derived from the rate of progress of the flame front (F_s) and another related to the rate of heat evolution by the material (Q) are combined to provide a flame spread index (I_s) or $I_s = F_s \times Q$. The composite panels were tested according to the procedure described previously (13). The panels were tested with the film side facing the radiant heat source. The test results are given in Fig. 10. The following observations were made during the testing of these panels:

1. Baseline Panel: Considerable charring, bubbling, and cracking occurred on the specimen surface. The panel core maintained good structural integrity and had slight smoke evolution.

2. Panel A: Considerable melting, bubbling, and shrinking occurred. The panel core maintained good structural integrity with moderate charring on the surface and had slight smoke evolution.

3. Panel B: Considerable melting and shrinking occurred. The panel core maintained good structural integrity and had very light smoke evolution. A disparity among the flame-spread indices is indicative of a much greater heat rise in the case of specimens 3 and 4, and, in the case of specimen 2, a flame-front advance which did not extend to the first data point at 7.5 cm.

4. Panel C: Considerable melting, shrinking, and bubbling occurred. The panel core maintained good structural integrity with moderate charring and slight flaking, and had slight smoke evolution.

5. Panel E: Considerable melting, bubbling, and shrinking occurred. The panel core maintained good structural integrity with slight charring and swelling and had slight smoke evolution. The higher flame spread indices for specimen 4 are due to a flame-spread advance which extended to the 15-cm data point.

6. Panel F: Considerable melting and shrinking occurred. The panel core maintained good structural integrity, with moderate charring, swelling, and blistering, and had moderate smoke evolution.

7. Panel G: Considerable bubbling of the facing noted shortly after radiant heat exposure. Surface flaming was confined to the facing material. The panel core maintained good structural integrity. A higher flame-spread index for specimen 1 is due to a greater heat rise.

8. Panel H: The panel was not tested because of the nonavailability of a large specimen.

Thermal analyses of the baseline and panel type B (XU71775/H795 Tape) were conducted on a DuPont 1090 thermogravimetric analyzer using both

nitrogen and air atmospheres with a sample size of 10 mg. The TGA data of 10°C/min heating rate in nitrogen and air are shown in Fig. 11. It can be seen that, under both the nitrogen and air environment, panel type B had a higher char yield than the baseline epoxy panel.

The mass loss of the composite panels was determined using the modified NBS Smoke Chamber described previously (14). At least three specimens were tested from each of the eight panel configurations at a radiant heat flux of 3.5 W/cm^2. The percent mass loss as a function of time is shown in Fig. 12. The percent mass loss is governed by this equation:

$$\text{\% Mass loss} = \frac{\text{Mass loss at any time interval}}{\text{Original mass}} \times 100 \qquad (5)$$

It can be seen that the panels fabricated with the PSP 6022 resin (panels type G and H) had the lowest weight loss of all the specimens tested. This was followed by the panel fabricated with the XU71775/H795 resin using the graphite unidirectional tape (panel type B). The panel with the highest weight loss was the baseline epoxy-fiberglass panel. Weight loss of the composite panel is an important parameter since it corresponds directly with the amount of visible smoke and other toxic gases produced during the pyrolysis of the sample. The lower rate of weight loss corresponds with a lower rate of pyrolysis products produced. In the mass loss test, a 6% difference in weight loss was observed between the baseline and panel type B (XU71775/H795/Tape/PEEK). In the TGA test a 6% difference was also observed between the two panels at 900°C in air, indicating a good correlation between the thermogravimetric analysis and the mass-loss test procedure just described. Figure 13 shows the effect of heating the epoxy-fiberglass composite and the XU71775/H795 graphite fabric composite. After heating for 5 min at 3.5-W/cm^2 input heat flux, the baseline epoxy-

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fiberglass composite was severely delaminated. The XU71775/H795 was almost intact as also shown by its low weight loss.

MECHANICAL PROPERTIES

The flexural strength and modulus, peel and tensile strength, and density of the composite panels were determined per MIL-STD-401 (15). The properties of the baseline epoxy-glass fabric panel, 2.54 cm thick, have been reported previously (16). The present panels tested were approximately 0.67 cm thick.

The flexural strength and modulus of the composites were measured using the sandwich beam flexure apparatus. The bottom span was 55 cm and the top span 10 cm. The test results are given in Fig. 14. Panels type B and D showed the highest compressive stress. Panels constructed with the unidirectional tape showed the highest flexural strength. Panel type B in a thickness of 0.72 cm had a compressive stress of 91.1 kg/cm^2 compared to 70.2 kg/cm^2 for a 2.54-cm-thick baseline epoxy-glass fabric panel.

The peel strength of the composites was determined using the climbing drum peel apparatus. The rate of test was 2.5 cm/min, the drum radius was 5.0 cm, the flange radius was 6.3 cm, and the torque arm was 1.2 cm long. The test results are given in Fig. 15. Panel type F had the highest peel strength. The baseline panel has a peel strength of 1.3 cm·kg/cm width (16).

The flatwise tensile strength of the panels is given in Fig. 16. As a comparison, the flatwise tensile strength of the baseline panel is 19.0 kg/cm^2.

The density of the panels is given in Table 6. Panel type B has approximately equivalent density to the baseline panel. All of the graphite fabric panels have a lower density than the baseline panel.

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CONCLUSIONS

To rank the composites, one should consider all of the materials parameters and assign weight to each specific parameter or measurement. Recent studies (10) have indicated that a low rate of heat release or fuel contribution is one of the most important parameters to be considered when using composites in critical applications such as aircraft. Based on these observations, the following conclusions may be drawn from this review:

1. The highest total-heat release and heat-release rates were measured with baseline epoxy composite panel. This panel also exhibited the highest smoke evolution, highest mass losses, highest CO evolution, and lowest oxygen index of all the composites tested.

2. The type B panel (XU71775/H795 Tape/PEEK) exhibited the lowest heat-release rate and total-heat release. It was the only panel tested which meets the performance criteria of maximum heat-release rate of 65 kW/m^2 and maximum total-heat release in 2 min of 65 kW·min/m^2. This composite panel measured the highest oxygen index of all the panels.

3. The lowest smoke evolution was measured in panel type G (PSP 6022M/Fabric/PEEK) and panel type D (H795 Tape/PEEK).

4. All graphite panels in the ignition resistance test were "self extinguishing" and panel type G exhibited the shortest burn length.

5. The panel type A (XU71775/H795/Fabric/PEEK) for the surface flammability test had the lowest flame spread index followed by panel type C (H795 Fabric/PEEK) and the baseline.

6. All the graphite composites exhibited oxygen indices significantly higher than the baseline panel, indicating that the graphite panels will exhibit lower relative flammability. Composite panels A, B, and D showed the highest indices (44.3, 45.6, 45.0, respectively), compared to 34.6 for the baseline panel.

7. At approximately equivalent densities, panel type B (XU71775/H795 Tape/PEEK) has much higher flexural and tensile strength than the baseline epoxy-fiberglass composite panel.

8. The aforementioned data indicate that composites fabricated with the XU71775/H795 vinyl polystyrylpyridine/mismaleimide resin exhibited the optimum combination of fire-resistant properties and processing characteristics.

LIST OF SYMBOLS

A = exposed-test-specimen surface area

D_{max} = specific optical density, maximum

D_s = specific optical density

dt = time interval

F_s = flame spread factor

HRR = heat-release rate

I_s = flame spread index

K_h = calibration factor (kW/mV)

L = length of light path

Plt = percent light transmission

Q = heat-evolution factor

R = resin content

T_i = inlet temperature

T_o = outlet temperature

t = time

V_b = "blank" thermopile voltage

V_m = measured thermopile voltage (mV)

V_o = volatile content

V_1 = chamber volume

V_2 = volume of air

w_f = weight of fibers

w_p = weight of prepreg

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BIOGRAPHY

Demetrius A. Kourtides is a research scientist in the Thermal Protection Materials Branch of NASA Ames Research Center, Moffett Field, California.

He received his M.S. Degree in Chemical Engineering in 1959 from Brigham Young University, Provo, Utah. From 1959 to 1965 he was an Associate Research Scientist at Varian Associates, Palo Alto, California. Since 1965 after joining NASA, he has done research in the area of thermally stable thermoset and thermoplastic polymers with special emphasis in the development of fire-resistant composites. He has authored ninety publications in this field and holds eight patents relating to composite structures and fire-resistant polymers and fabrics. He is an active member of the Society of Plastics Engineers and has served as Technical Program Chairman. He is also a member of the American Chemical Society and SAMPE.

In addition he has written two review articles in professional journals. His awards include "Best of Issue" award issued by the Journal of Plastics Design and Processing, the Congressional Excalibur Award in 1984 for excellence in public service, and the NASA Space Act Award in 1985 for the development of fire-resistant fabrics.

Table 1

PANEL TYPE	CURE	POST-CURE
C: H795 FABRIC/PEEK D: H795 TAPE/PEEK	AUTOCLAVE, VACUUM 584 mm Hg, PRESSURE 690 kPa 1.5°C/min, 177°C, 2 hr COOL TO 66°C	10°C/min, 204°C, 1 hr 1.5°C/min, 232°C, 10 min COOL TO 66°C BOND TO HONEYCOMB WITH FILM ADHESIVE 1.5°C/min, 121°C, 30 min 1.5°C/min, 177°C, 4 hr COOL TO 66°C
G: PSP6022 FABRIC/PEEK H: PSP6022 TAPE/PEEK	1.5°C/min, 177°C, 1 hr, 1.5°C/min, 204°C 15 min AUTOCLAVE, VACUUM 584 mm Hg, 10 min PRESSURE 1034 kPa AT 69 kPa/min VENT VACUUM AT 139 kPa 1034 kPa, 204°C, 9 hr COOL TO 66°C VENT	10°C/min, 249°C, 2 hr COOL TO 66°C BOND TO HONEYCOMB SAME AS ABOVE

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Table 2

PANEL TYPE	CURE	POST-CURE
A: XU71775/H795 FABRIC/ PEEK	AUTOCLAVE, VACUUM 584 mm Hg, PRESSURE 173 kPa 1.5°C/min, 130°C, 20 min	1.5°C/min, 177°C, 18 hr
B: XU71775/H795 TAPE/ PEEK	1.5°C/min, 177°C, 6 hr COOL TO 66°C RELEASE PRESSURE	
E: CYCOM 6162 FABRIC/ PEEK	AUTOCLAVE, VACUUM 584 mm Hg, PRESSURE 173 kPa 1.5°C/min, 132°C 1 hr	1.5°C/min, 177°C, 8 hr
F: CYCOM 6162 TAPE/ PEEK	COOL TO 66°C RELEASE PRESSURE	

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Table 3

PANEL TYPE	OXYGEN INDEX
BASELINE EPOXY GLASS FABRIC/PVF	34.6
A-XU71775/H795 FABRIC/PEEK	44.3
B-XU71775/H795 TAPE/PEEK	45.6
C-H795/FABRIC/PEEK	35.7
D-H795 TAPE/PEEK	45.0
E-CYCOM 6162/FABRIC/PEEK	38.8
F-CYCOM 6162/TAPE/PEEK	36.9
G-PSP 6022M/FABRIC/PEEK	40.3

Table 4

GAS CONCENTRATION, ppm [a]	PANEL TYPE							
	BASELINE	A	B	C	D	E	F	G
CO	200-700	75-300	50-400	100-400	100-400	100-325	50-300	100-300
NO_x	5-20	5-20	2-15	5-20	15-20	TRACE-5	15-20	0-10
HF	0	0	0	0	0	0	0	0
HCN	2-10	5-10	5-10	2-10	3-15	5	2-13	2-5

[a] NOTE: Values given are the high and low values obtained on three samples.

KOURTIDES

Table 5

TEST PARAMETER	PANEL TYPE						
	BASELINE	A	B	C	E	F	G
AVERAGE BURN LENGTH, cm (in.)	12.55 (4.94)	15.72 (6.19)	14.94 (5.88)	13.82 (5.44)	12.55 (4.94)	11.43 (4.5)	6.05 (2.38)
AFTERFLAME, sec	10.5	0.0	0.0	0.0	0.0	0.0	0.0

[a] There was no drip burn or afterglow.

KOURTIDES

Table 6

PANEL TYPE	DENSITY, g/cm^3 (lb/ft^3)	UNIT WT., kg/m^2 (lb/ft^2)
BASE-LINE	0.283 (17.36)	1.790 (0.365)
A	0.196 (12.23)	1.226 (0.250)
B	0.277 (17.29)	1.742 (0.357)
C	0.222 (13.85)	1.399 (0.287)
D	0.318 (19.84)	2.240 (0.459)
E	0.183 (11.41)	1.148 (0.235)
F	0.315 (19.65)	1.978 (0.405)
G	0.210 (13.10)	1.289 (0.264)
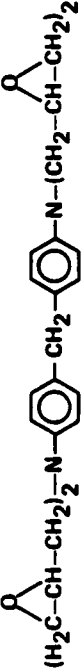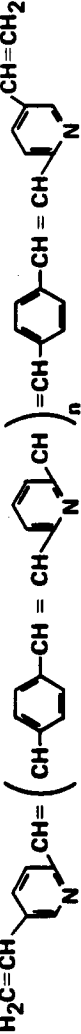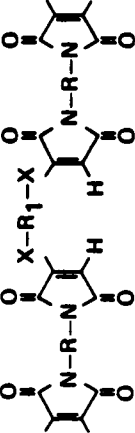
RESIN	DESIGNATION	COMPOSITE	CHEMICAL STRUCTURE
EPOXY	MXB 7203	BASELINE: EPOXY/GLASS FABRIC	$(H_2C-CH-CH_2)_2-N-\bigcirc-CH_2-\bigcirc-N-(CH_2-CH-CH_2)_2$
3 VINYLPOLYSTYRYL-PYRIDINE/MALEIMIDE	XU 71775/H795	A: XU 71775/H795 FABRIC B: XU 71775/H795 TAPE	(vinylpolystyrylpyridine structure, $H_2C=CH \ldots (CH \ldots CH=CH)_n \ldots CH=CH_2$)
BISMALEIMIDE	H795	C: H795/FABRIC D: H795/TAPE	(bismaleimide structure, $X-R_1-X$, $N-R-N$)

Fig. 1a

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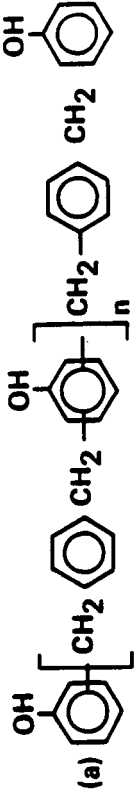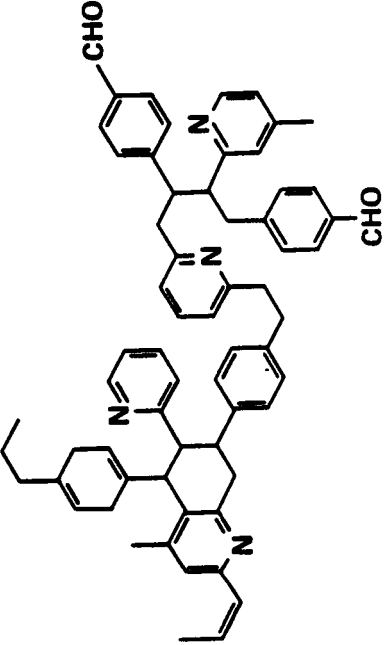
RESIN	DESIGNATION	COMPOSITE	CHEMICAL STRUCTURE
PHENOLIC	CYCOM 6162	E: CYCOM 6162/ FABRIC F: CYCOM 6162/ TAPE	
POLYSTYRYL-PYRIDINE	PSP6022M	G: PSP6022/ FABRIC H: PSP6022/ TAPE	

Fig. 1b

Fig. 2

Fig. 3

PEEK FILM

ADHESIVE

PREPREG (AS-4 1 PLY FABRIC
OR 3 PLY 0°90°0° TAPE)

CORE

PREPREG

Fig. 4

(FLAMING)

△ BASELINE
● A—XU71775/H795 FABRIC/PEEK
○ B—XU71775/H795 TAPE/PEEK
□ C—H795 FABRIC/PEEK

Fig. 5a

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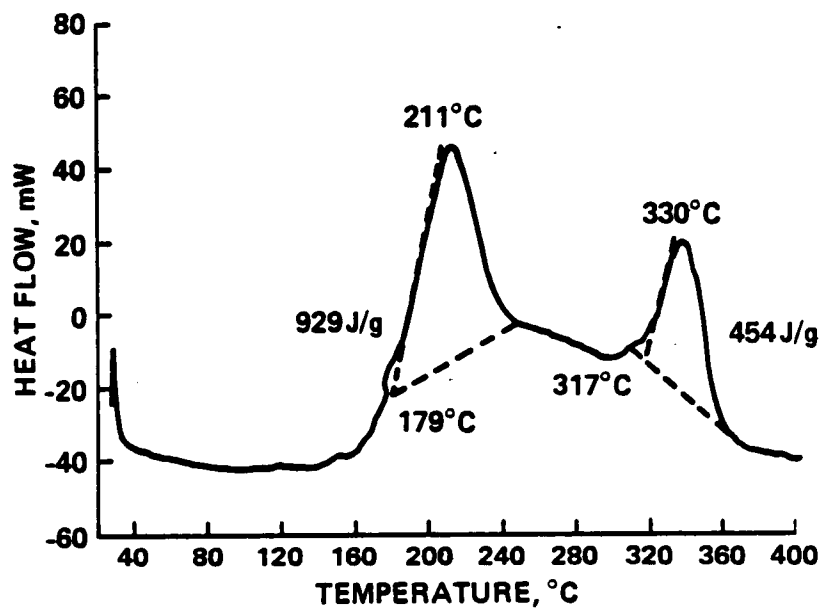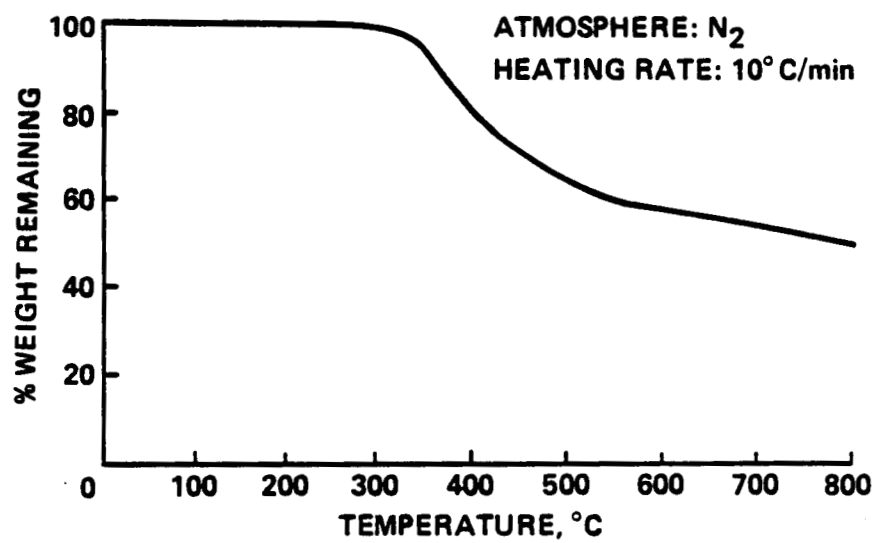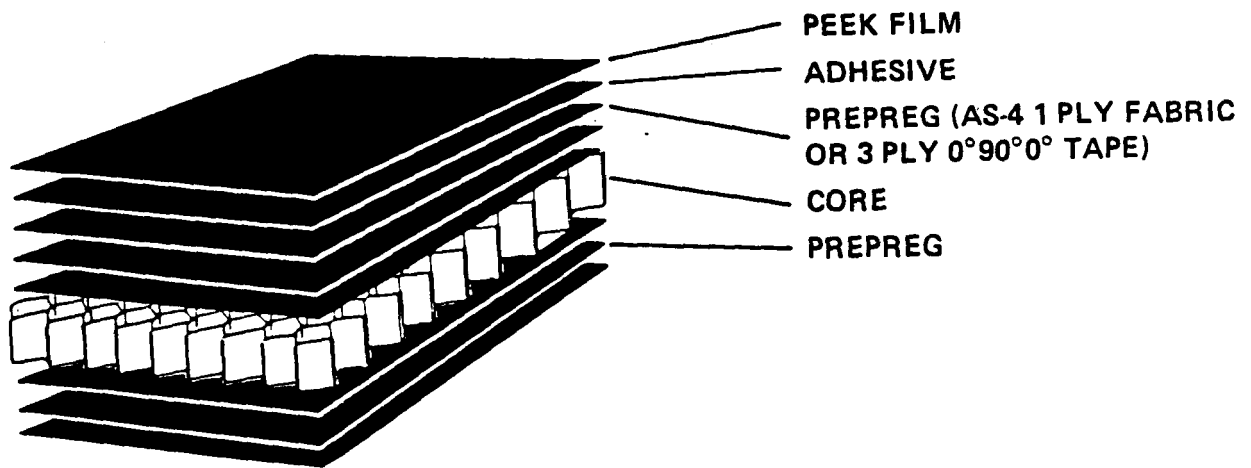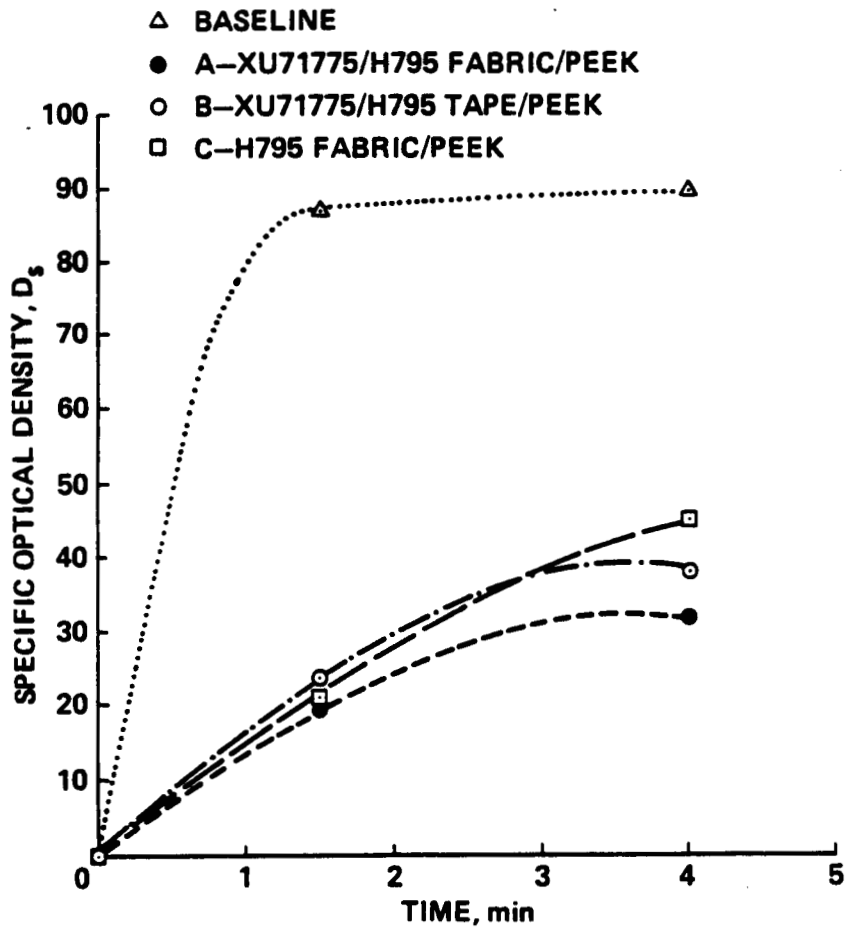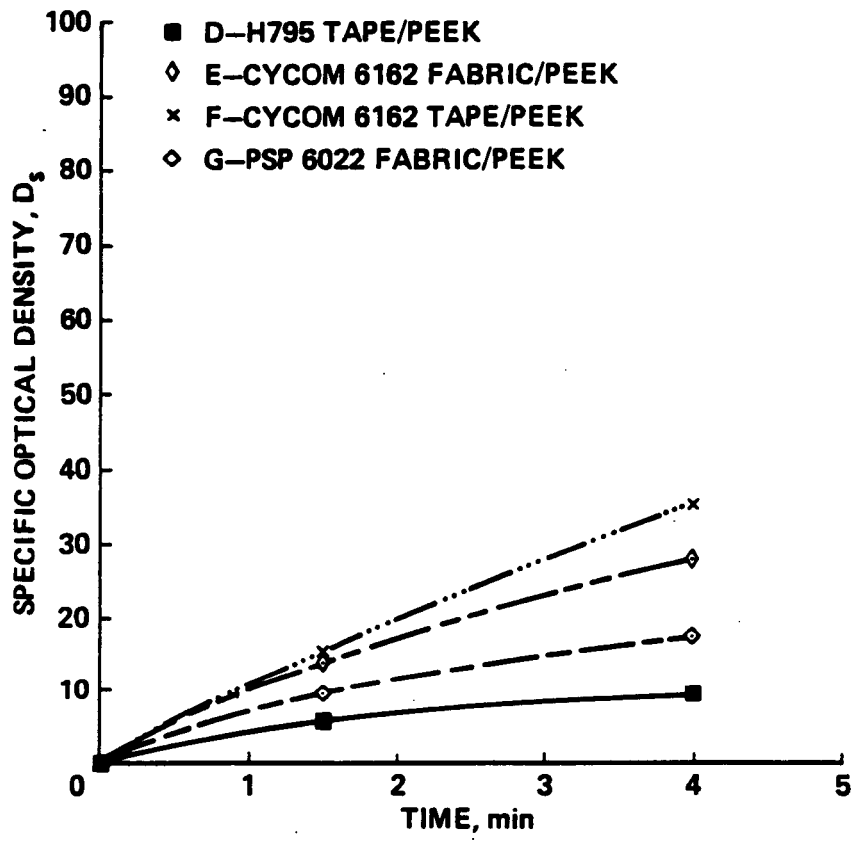
(FLAMING)
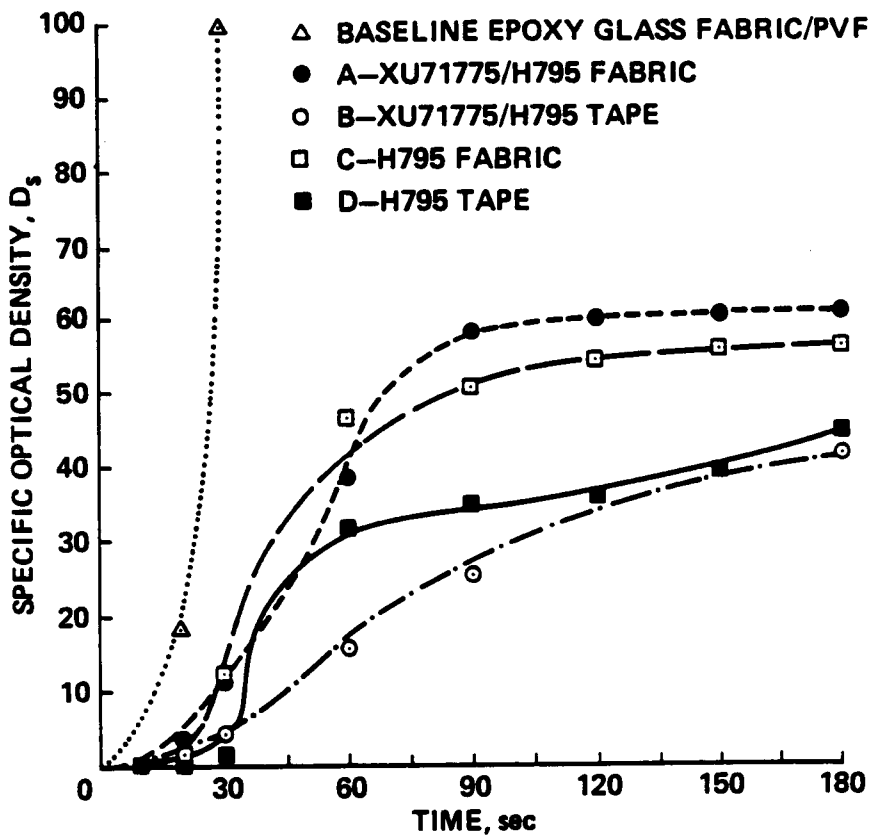
Fig. 5b
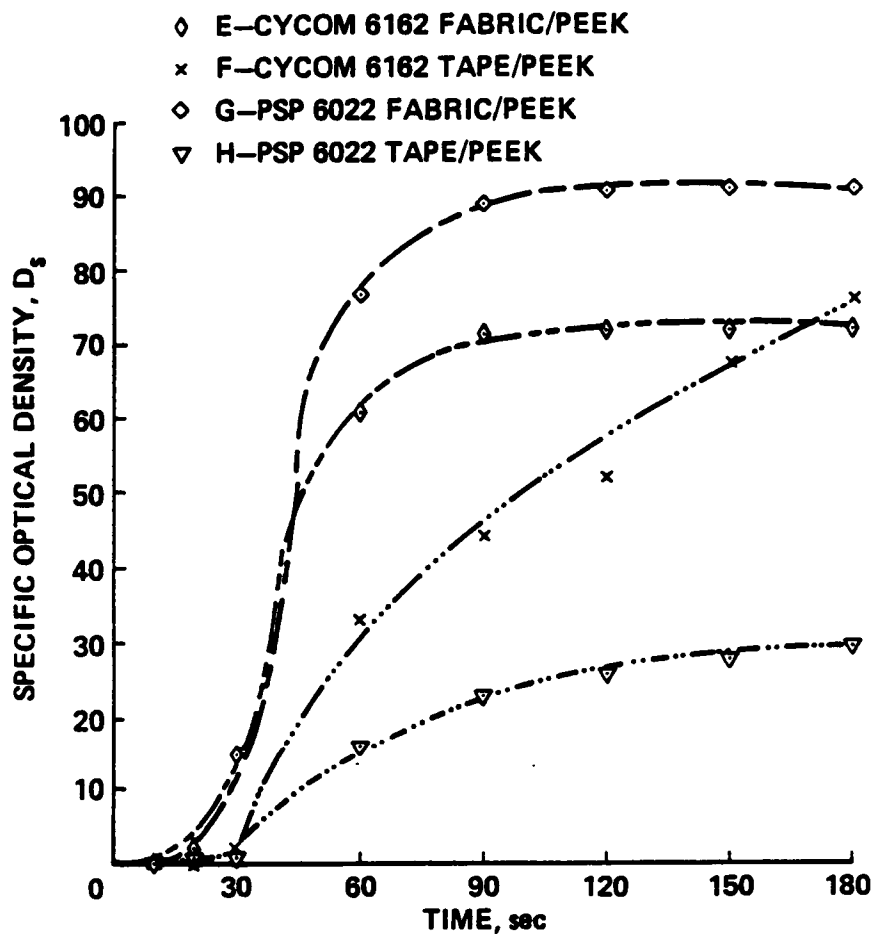
(NONFLAMING)
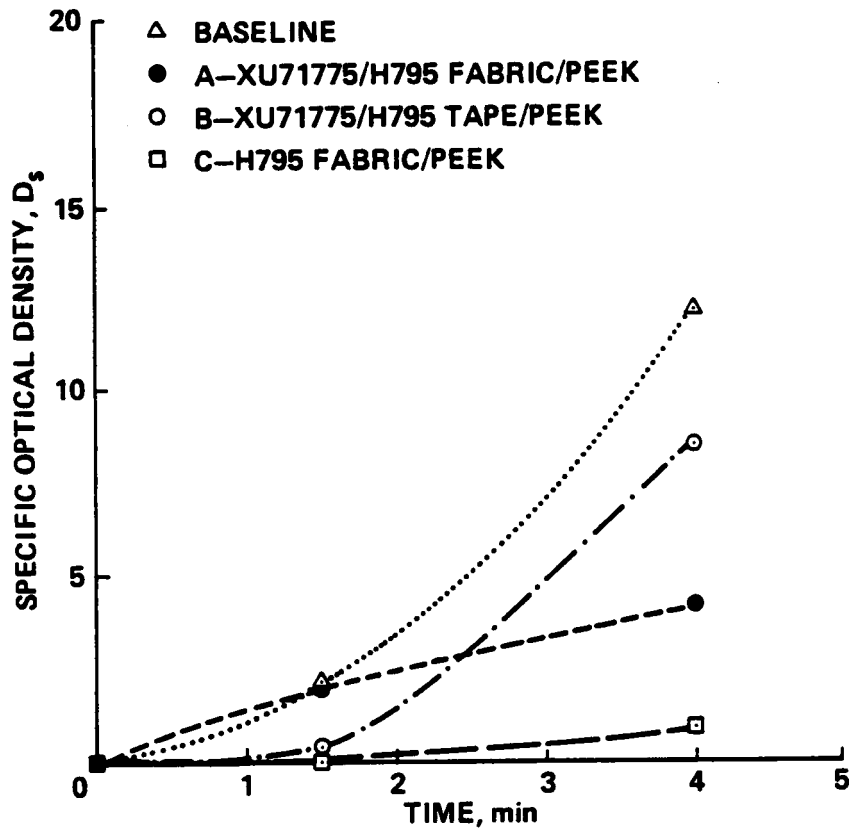
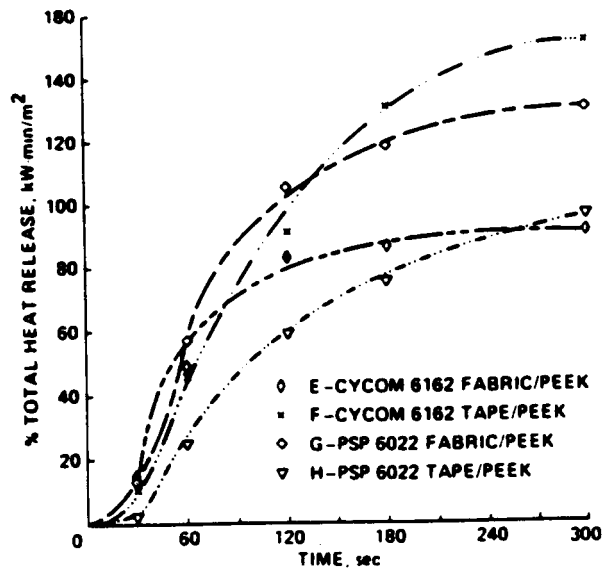Fig. 6a

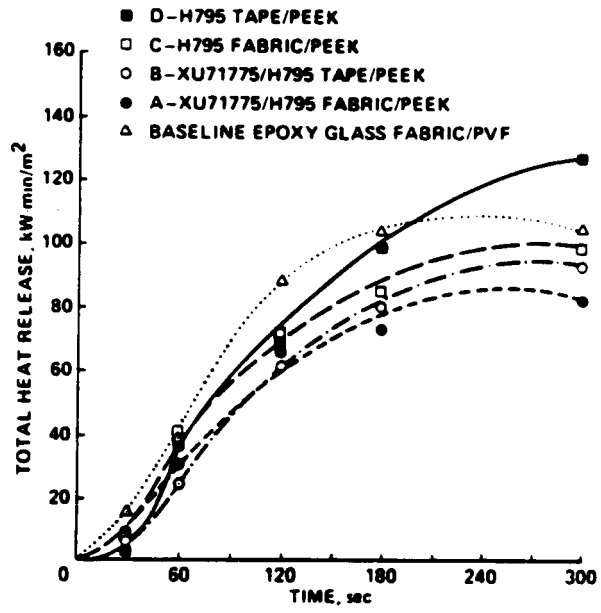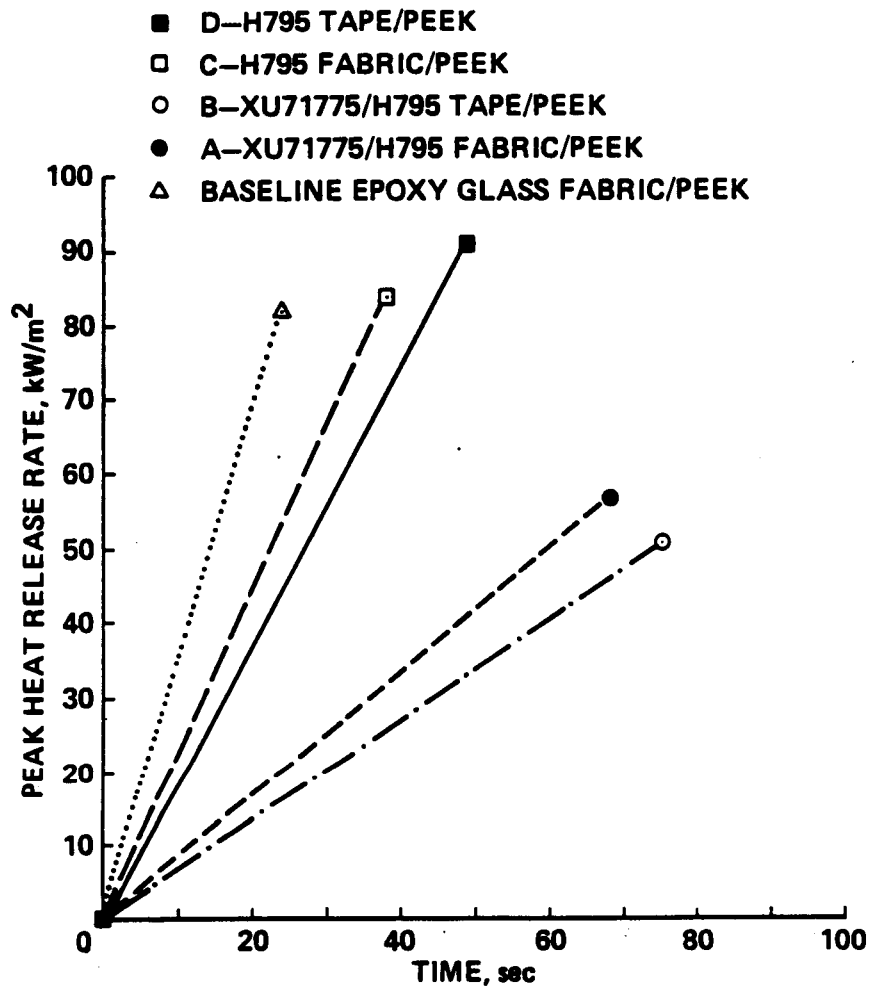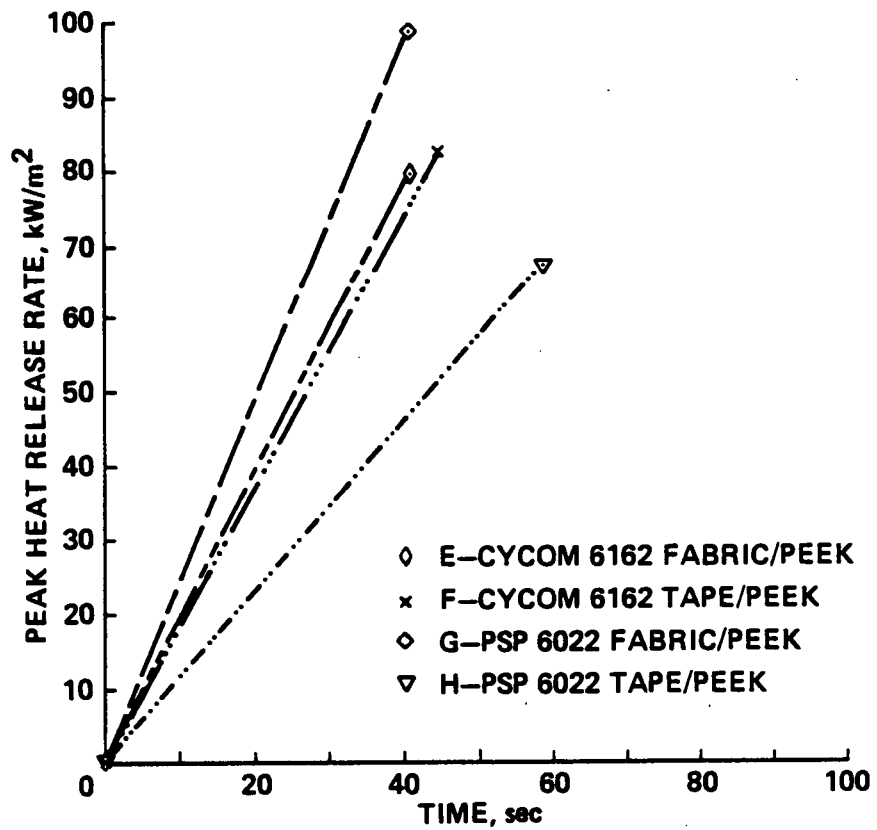**(NONFLAMING)**

Fig. 6b

Fig. 7a

38

Fig. 7b

39

Fig. 8

Fig. 9a

Fig. 9b

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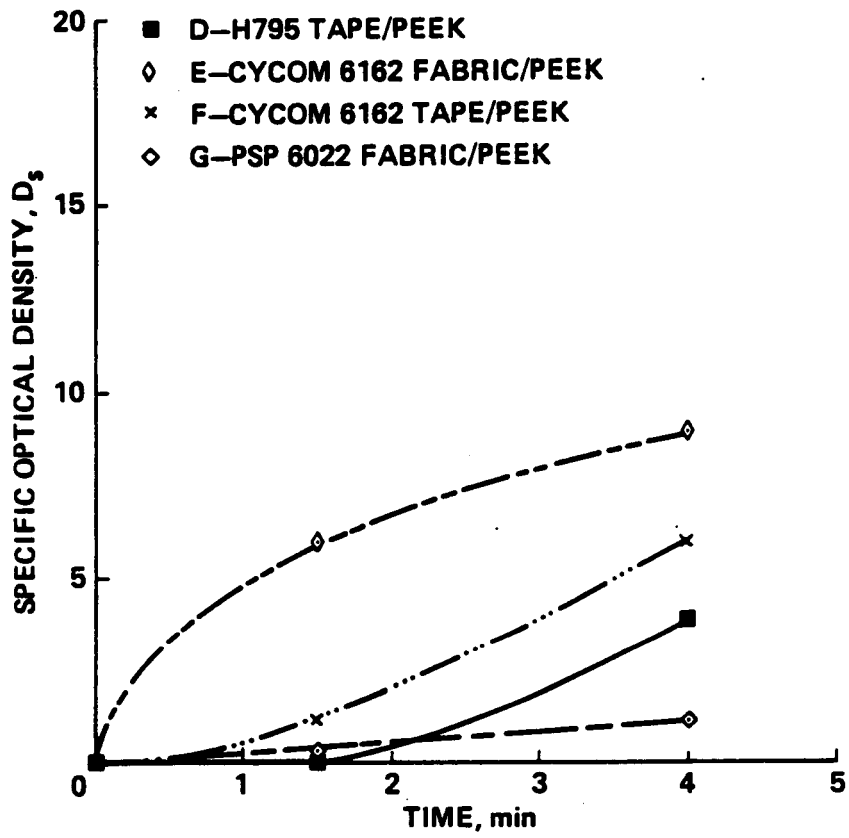
FLAME SPREAD INDEX OF COMPOSITES

Fig. 10

■ XU71775/H795 (IN NITROGEN)
□ BASELINE EPOXY (IN NITROGEN)
○ XU71775/H795 (IN AIR)
● BASELINE EPOXY (IN AIR)

Fig. 11

Legend:
- ■ D—H795 TAPE/PEEK
- □ C—H795 FABRIC/PEEK
- O B—XU71775/H795 TAPE/PEEK
- ● A—XU71775/H795 FABRIC/PEEK
- △ BASELINE EPOXY GLASS FABRIC/PEEK
- ◊ E—CYCOM 6162 FABRIC/PEEK
- × F—CYCOM 6162 TAPE/PEEK
- ◇ G—PSP 6022 FABRIC/PEEK
- ▽ H—PSP 6022 TAPE/PEEK

$3.5 \ W/cm^2$

Y-axis: % MASS LOSS
X-axis: TIME, sec

Fig. 12

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Front

Back

Front

Back

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FLEXURAL STRENGTH, kg/cm

BASELINE:
EPOXY GLASS FABRIC

A:
XU71775/H795
FABRIC/PEEK

B:
XU71775/H795
TAPE/PEEK

C:
H795 FABRIC/PEEK

D:
H795 TAPE/PEEK

E:
CYCOM 6162 FABRIC/PEEK

F:
CYCOM 6162 TAPE/PEEK

G:
PSP 6022 FABRIC/PEEK

Fig. 14

PEEL STRENGTH, cm-kg/cm WIDTH

BASELINE:
EPOXY GLASS FABRIC

A:
XU71775/H795
FABRIC/PEEK

B:
XU71775/H795
TAPE/PEEK

C:
H795 FABRIC/PEEK

D:
H795 TAPE/PEEK

E:
CYCOM 6162
FABRIC/PEEK

F:
CYCOM 6162
TAPE/PEEK

G:
PSP 6022 FABRIC/PEEK

Fig. 15

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FLATWISE TENSILE STRENGTH, kg/cm^2

BASELINE:
EPOXY GLASS FABRIC

A:
XU71775/H795
FABRIC/PEEK

B:
XU71775/H795
TAPE/PEEK

C:
H795 FABRIC/PEEK

D:
H795 TAPE/PEEK

E:
CYCOM 6162
FABRIC/PEEK

F:
CYCOM 6162
TAPE/PEEK

G:
PSP 6062
FABRIC/PEEK

Fig. 16

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16. Abstract

Flammability, thermal, and selected mechanical properties of composites fabricated with epoxy and other thermally stable resin matrices are described. Properties which were measured included limiting-oxygen index, smoke evolution, thermal degradation products, total-heat release, heat-release rates, mass loss, flame spread, ignition resistance, thermogravimetric analysis, and selected mechanical properties. This paper describes the properties of eight different graphite composite panels fabricated using four different resin matrices and two types of graphite reinforcement. The resin matrices included: XU71775/H795, a blend of vinylpolystyrylpyridine and bismaleimide; H795, a bismaleimide; Cycom 6162, a phenolic; and PSP 6022M, a polystyrylpyridine. The graphite fiber used was AS-4 in the form of either tape or fabric. The properties of these composites were compared with epoxy composites. It was determined that the blend of vinylpolystyrylpyridine and bismaleimide (XU71775/H795) with the graphite tape was the optimum design giving the lowest heat release rate.

17. Key Words (Suggested by Author(s)) Graphite composites Thermal properties Flammability	18. Distribution Statement Unclassified-Unlimited Subject Category - 24

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