POLYMER RESEARCH
AT NASA LANGLEY RESEARCH CENTER

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INTRODUCTION

The polymer program at NASA Langley Research Center has as a focus the synthesis and characterization of polymers for aerospace applications. Requirements for these materials vary according to the specific programs.

The synthesis effort involves preparation of polymers for both intermediate- and high-temperature applications. The systems under investigation are thermoplastics, thermosets, and hybrids of these two.

The characterization effort includes a wide variety of programs, such as methodology development, general testing, and specialized studies. This work deals with the elucidation of polymer behavior in composite, adhesive, and film applications for the various aerospace applications.

FOCUS

- Polymer Synthesis
  - Thermoplastics
  - Pseudothermoplastics
  - Thermosets
- Characterization
  - Methodology Development
  - General Testing
  - Specialized Studies

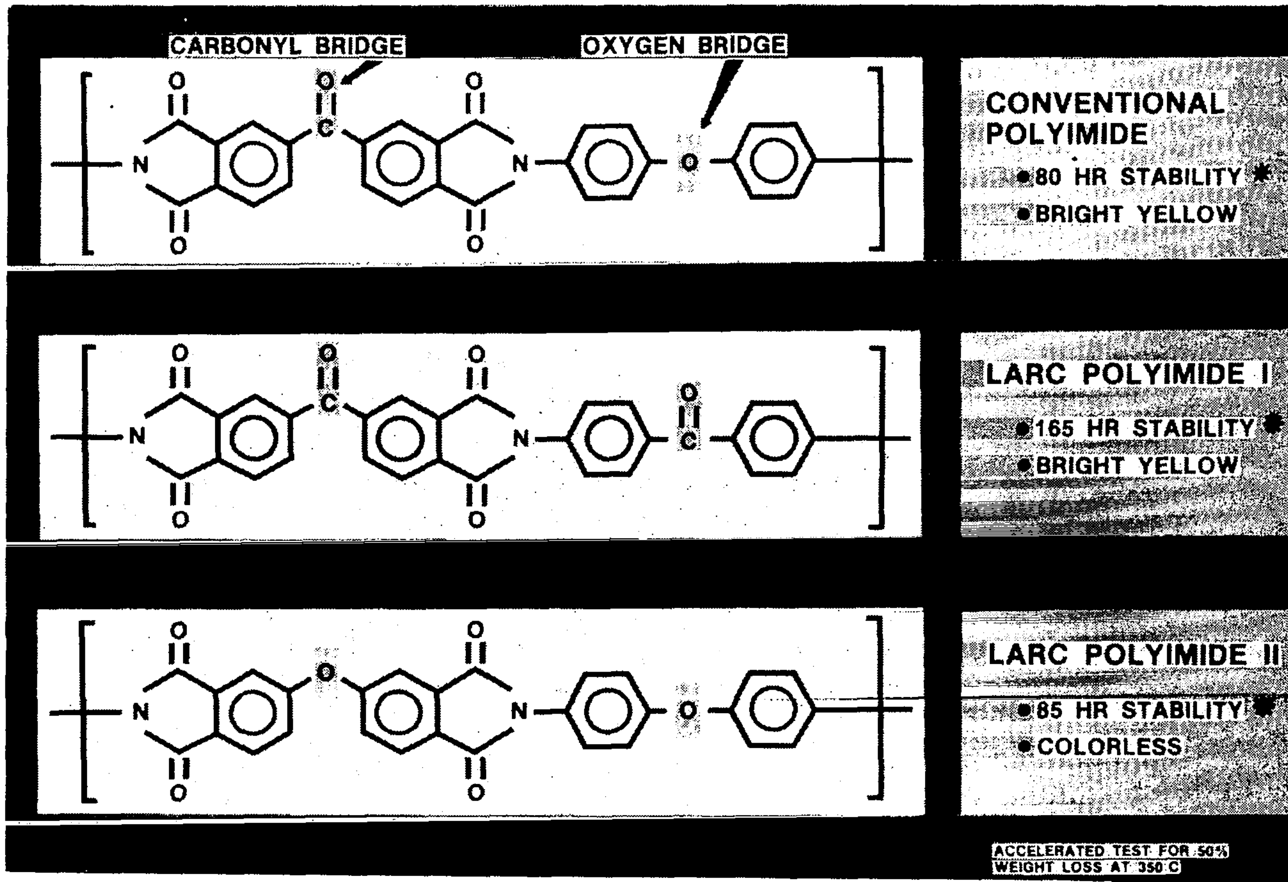
POTENTIAL APPLICATIONS

- Matrix Resins
- Adhesives
- High Performance Films

TAILORING POLYMER STRUCTURES
TO CONTROL PROPERTIES

The oxidative stability and color (transparency) of linear aromatic polymers can be tailored by altering chemical groups that bridge the aromatic rings. In the figure below, the $-\overset{\text{O}}{\overset{\|}{\text{C}}}-$ (carbonyl) bridging group is in the dianhydride-derived portion of the polymer and the -o- (oxygen) bridging group is in the diamine-derived portion of the commercial polyimide. When this polyimide was tested at 350°C the polymer film lost half of its initial weight in 80 hours. Other experimental polyimides were prepared and tested in the same manner, but the bridging groups were altered as shown in the figure. With the carbonyl bridge in both components (LARC Polyimide I), oxidative stability was improved by a factor of 2 without affecting the color transparency. Altering the structure by placing the oxygen bridge in both components (LARC Polyimide II) provided a polymer film with good transparency (colorless) and oxidative stability comparable to the commercial polyimide.

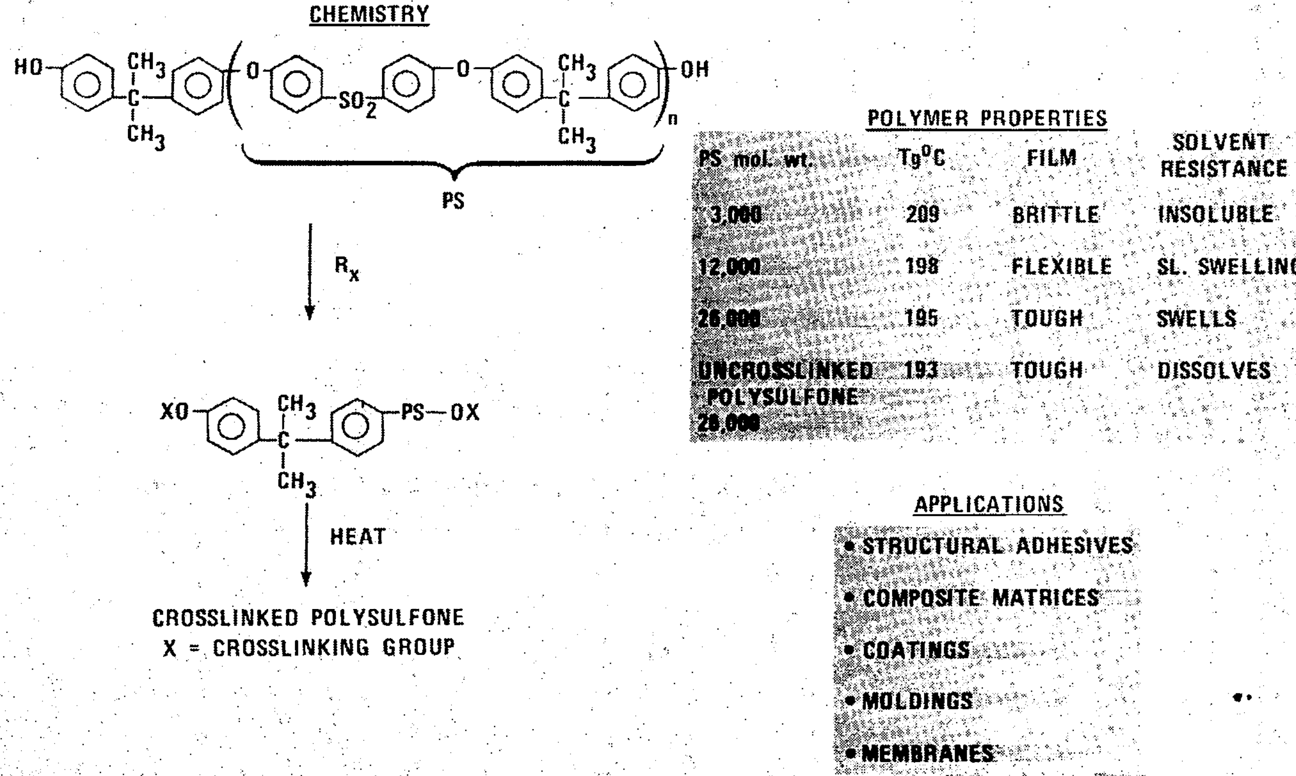
These results indicate the progress we are making in understanding how to control properties by tailoring the chemical structure of the polymer. For polymer coatings we can achieve good transparency without sacrificing oxidative stability, or the structure can be altered to achieve excellent gains in thermooxidative stability (ref. 1).



TOUGH SOLVENT RESISTANT POLYSULFONES

Polysulfones are engineering thermoplastics which are widely used in a variety of applications. They exhibit an excellent combination of processability, mechanical properties including impact strength, and cost. Their use, however, is generally restricted to environments where there is no exposure to polar organic solvents. Polysulfones are prone to solvent attack, especially in a stressed condition, undergoing solvent-induced crazing and cracking. As a result their use as structural adhesives and composite matrices on aerospace vehicles has been restricted.

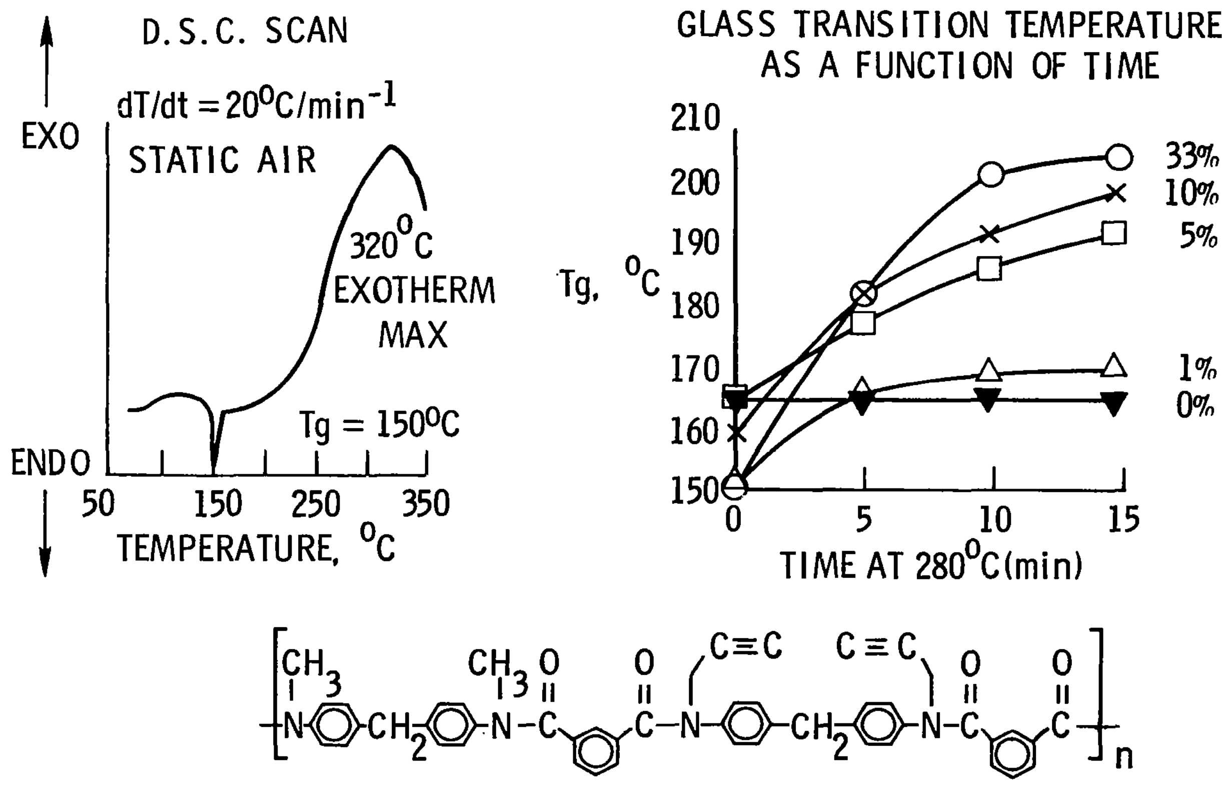
Several synthetic routes are under investigation to transform polysulfones into solvent resistant materials while retaining their attractive properties. As shown in the figure, cured polymers from polysulfones endcapped with crosslinkable groups exhibit better solvent resistance and higher use temperatures than commercial polysulfone. As the solvent resistance of the polymer is improved, there is a corresponding loss of toughness. Acceptable trade-offs can be made by adjusting the molecular weight to maximize the attractive features without severely compromising other properties (e.g., toughness and thermoformability). This approach, along with other routes, offers the promise of providing polysulfones with improved solvent resistance, thereby making them more acceptable for use as structural adhesives and composite matrices on airplanes (ref. 2).



N-PROPARGYL-SUBSTITUTED AROMATIC POLYAMIDES

A study was conducted to crosslink methyl-substituted polyamides via pendent propargyl groups for the purpose of improving the applicability of this resin as a matrix for Kevlar fiber composites. Films of the polyamides containing 1-33% propargyl diamine were successfully crosslinked by heating in air at 280°-300°C. The thermal crosslinking of the latent propargyl groups was evidenced by a rise in the glass transition temperature of the films with increasing propargyl concentration, a loss in solubility, and the disappearance of propargyl-related peaks from the infrared film spectra. Thermal crosslinking was accomplished with a slight loss in thermooxidative stability.

From the results of this investigation, propargyl-containing polyamides have been determined to be feasible matrices for polyamide fiber (Kevlar) composites. These materials are processable at advantageously low temperatures and are thermally crosslinkable at temperatures below the relaxation temperature of the fiber. Compatibility of these resins with aromatic polyamide fibers gives them high potential for success as matrix resins for Kevlar composites (ref. 3).

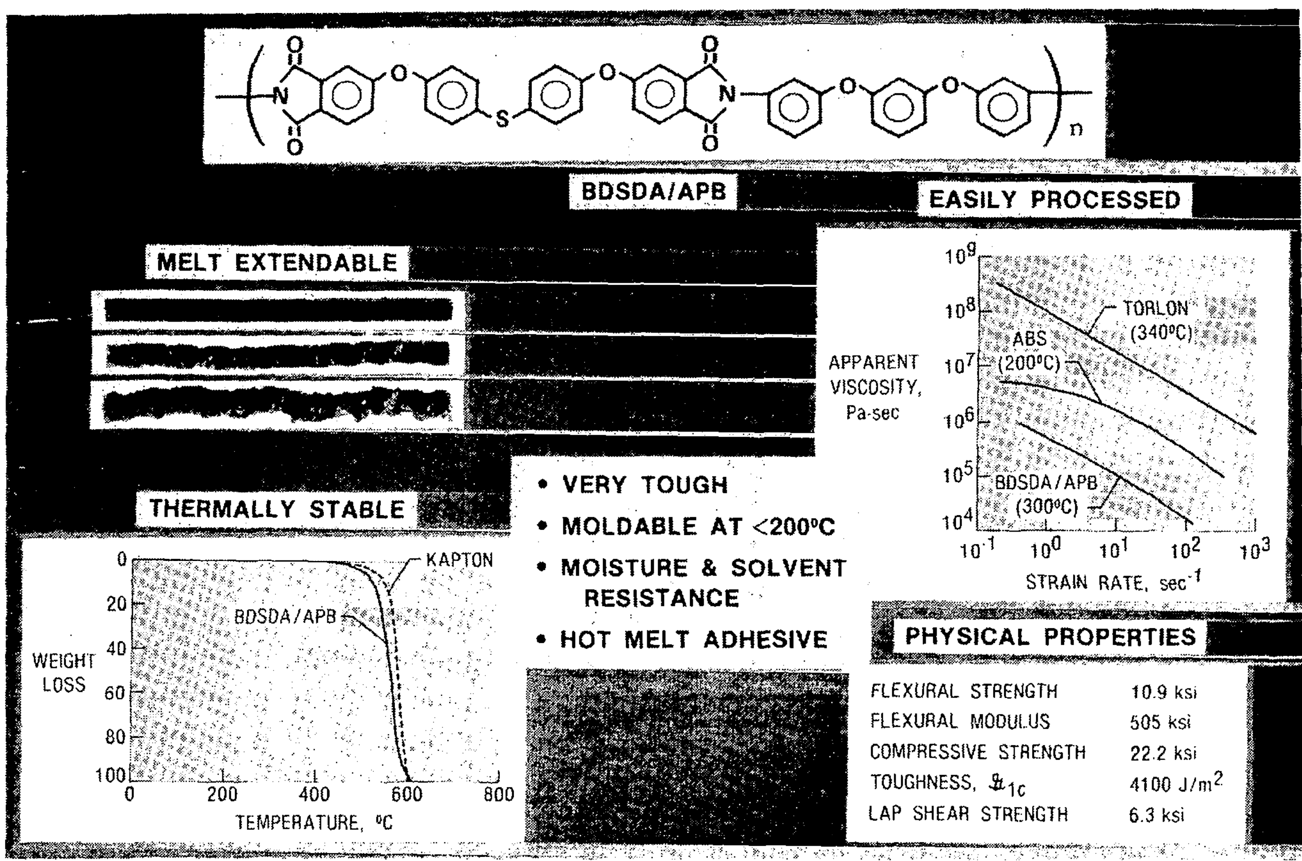


HOT MELT PROCESSABLE POLYIMIDE

Linear aromatic polyimides are a class of polymers which are generally not processable via conventional thermoplastic or hot-melt techniques. This class of polymer is, however, exceptionally thermally stable and has high glass transition temperatures. It is also resistant to attack by common organic solvents.

Linear aromatic polyphenylene oxides and sulfides, on the other hand, are more easily processed than the polyimides, generally exhibit lower glass transition temperatures, and still have relatively good thermal stability, although not equal to the polyimides. These systems also do not possess solvent resistance equal to the polyimides.

A novel linear aromatic polyphenylene ethersulfideimide has been synthesized which has some of the favorable characteristics of each parent system. The polymer has been molded, used as a resin, and cast into thin films. A limited characterization indicates this system can be processed via conventional thermoplastic techniques and may have a wide variety of applications (ref. 4).



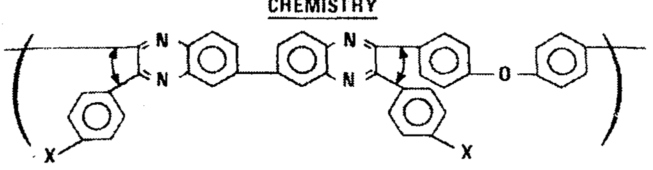
CROSSLINKING OF POLYPHENYLQUINOXALINES

In an attempt to overcome the shortcomings of thermoplastics without severely compromising their attractive features, work was initiated using polyphenylquinoxalines (PPQs) as a model system to demonstrate a general concept. Latent pendent groups (e.g. ethynyl and phenylethynyl) were incorporated on the linear molecules. These groups undergo a thermally induced reaction to provide controlled crosslinking. In this way, molecules are tied together (see figure) such that there is a significant improvement in their elevated temperature performance (e.g. creep resistance) and, more importantly, in their fluid and solvent resistance.

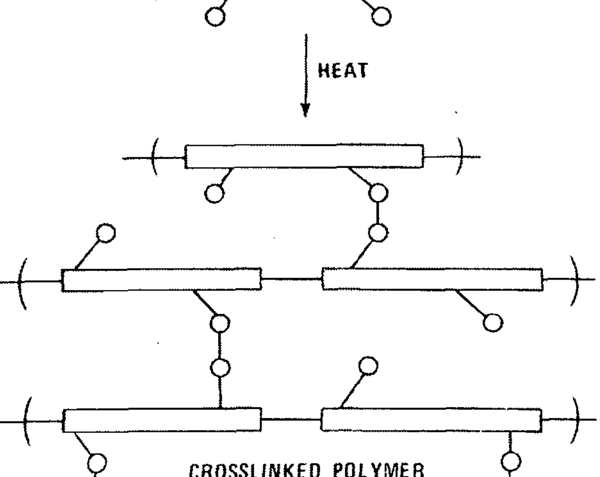
As indicated in the table, the T_g of a cured PPQ containing 5% of ethynyl (C≡CH) or phenylethynyl (C≡C-ϕ) groups was substantially higher than the parent PPQ void of latent crosslinking groups. At the 10% pendent group level, the cured PPQ containing the crosslinking groups became totally insoluble. Preliminary adhesive evaluation has also shown a marked increase in the 288°C lap shear strength. This concept is now being extended to polysulfones, which are lower temperature thermoplastics than PPQs, and it appears to be applicable to other thermoplastics such as polyesters, polyamides, and polyimides. This novel route offers the potential of modifying existing thermoplastics, particularly polysulfones, to improve their performance and make them acceptable for structural uses on future aircraft and spacecraft (ref. 5).

OBJECTIVE: DEVELOP SOLVENT RESISTANT HIGH TEMPERATURE STRUCTURAL RESINS

CHEMISTRY



LINEAR SOLUBLE POLYPHENYLQUINOXALINE
WHERE SOME X GROUPS ARE CROSSLINKERS



PROPERTIES

% OF CROSSLINKER	SOLVENT RESISTANCE	288°C LAP SHEAR STRENGTH, psi
0	DISSOLVES	1000
5	SWELLS	1800
10	INSOLUBLE	2200

APPLICATIONS

- HIGH TEMPERATURE ADHESIVES
- PROTECTIVE COATINGS
- HIGH TEMPERATURE COMPOSITE MATRICES
- ELECTRONIC COMPONENTS

ACETYLENE-TERMINATED PHENYLQUINOXALINES

A series of acetylene-terminated phenylquinoxaline (ATPQ) oligomers of various molecular weights were prepared and subsequently chain-extended by the thermally induced reaction of the ethynyl groups. The processability and thermal properties of these oligomers and their cured resins were compared with those of a relatively high molecular weight linear polyphenylquinoxaline (PPQ) with the same chemical backbone. The ATPQ oligomers exhibited significantly better processability than the linear PPQ but the PPQ displayed substantially better thermooxidative stability. Adhesive (Ti/Ti) and composite (graphite filament reinforcement) work was performed to evaluate the potential of these materials for structural applications. The PPQ exhibited better retention of adhesive and laminate properties than the ATPQ resins at 260°C after aging for 500 hr at 260°C in circulating air (ref. 6).

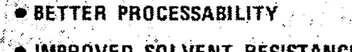
OBJECTIVE: IMPROVE THE PROCESSABILITY AND HIGH TEMPERATURE PERFORMANCE OF PPQ'S

HC≡C ... C≡CH

HEAT

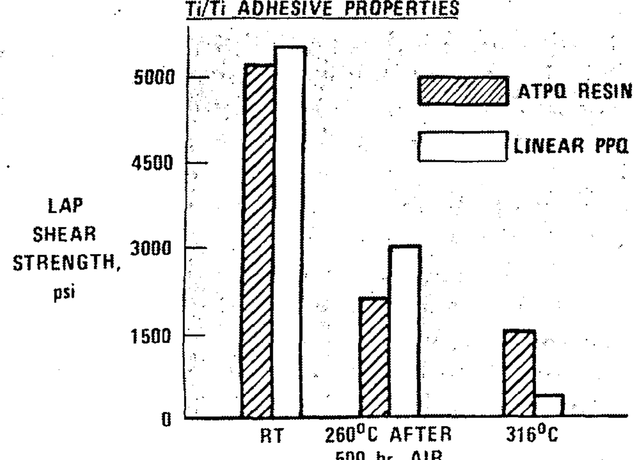
CROSSLINKED POLYMER (ATPQ RESIN)

ADVANTAGES OVER EXISTING POLYMERS



- HIGHER INITIAL ELEVATED TEMPERATURE STRENGTH
- AMENABLE TO TAILORING FOR SPECIFIC USE

Ti/Ti ADHESIVE PROPERTIES



HT-S UNIDIRECTIONAL LAMINATE PROPERTIES

TEST CONDITION	FLEXURAL STRENGTH, Ksi	FLEXURAL MODULUS, Msi
RT	237	19.3
260°C	201	17.1
260°C AFTER 500 hrs, AIR	189	16.8

ADDITION POLYIMIDE ADHESIVES

Addition polyimide oligomers have been synthesized from 3,3',4,4'-benzophenone tetracarboxylic acid dianhydride and 3,3'-methylenedianiline using a variety of latent crosslinking groups as end caps. The nominal 1300-molecular-weight imide prepolymers were isolated and characterized for solubility in amide, chlorinated, and ether solvents; melt-flow and cure properties; glass transition temperature; and thermal stability on heating in an air atmosphere. The general structure of the prepolymer and the end caps is shown below. Adhesive strengths of the polyimides were obtained both at ambient and elevated temperatures before and after aging at 232°C. Properties of the novel addition polyimides were compared to a known nadic end-capped adhesive, LARC-13 (ref. 7).

END GROUP
R

NADIC (LARC-13)

CH_3-NADIC

Cl_6-NADIC

ACETYLENE

N-PROPARGYL

CYCLOHEXENE

MALEIC

LAP SHEAR STRENGTHS (LSS) OF ADDITION POLYIMIDES AGED FOR 1000 HOURS AT 232°

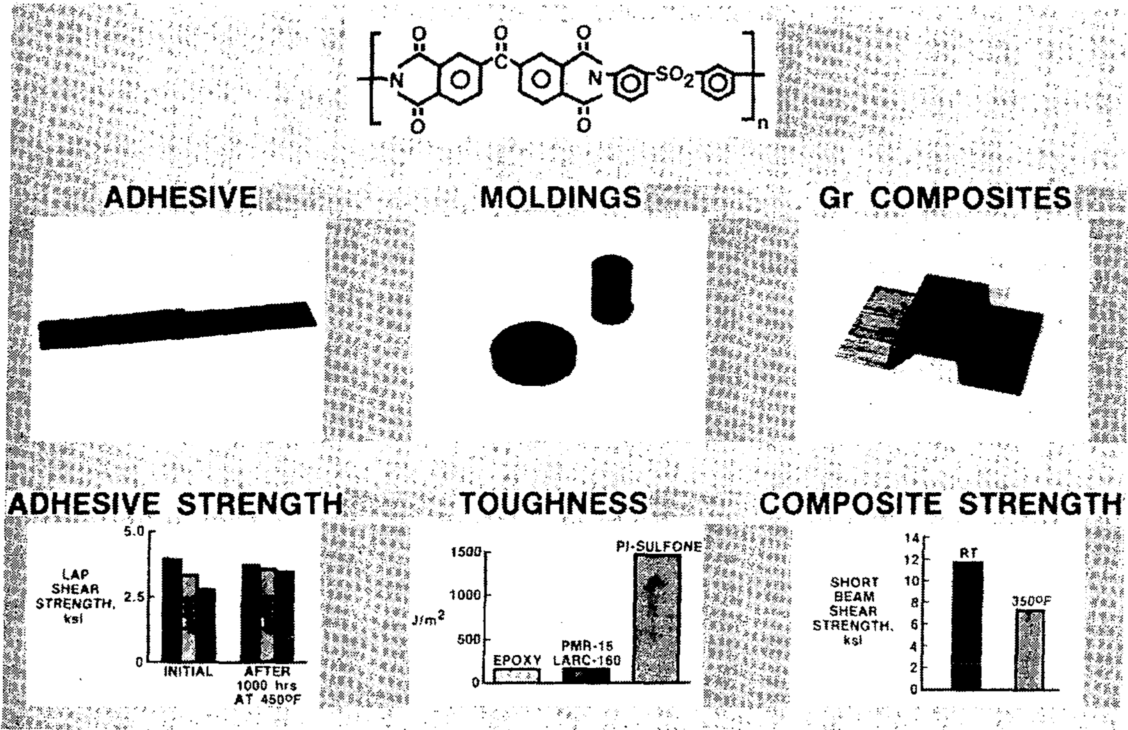
OLIGOMER END GROUP	INITIAL LSS, psi		LSS AFTER 1000 hr AT 232°C, psi	
	RT	232°C	RT	232°C
NADIC (LARC-13)	3200	2600	2600	1960
ACETYLENE	2900	2500	2500	2800
N-PROPARGYL	3100	2800	800	1000

THERMOPLASTIC POLYIMIDESULFONE

Aromatic polysulfones, a class of high-temperature engineering thermoplastics, have a major deficiency in their tendency to swell and dissolve in many common solvents. This solvation can cause structural components which are fabricated from these polymers to be susceptible to damage by these solvents and thereby lose their structural integrity.

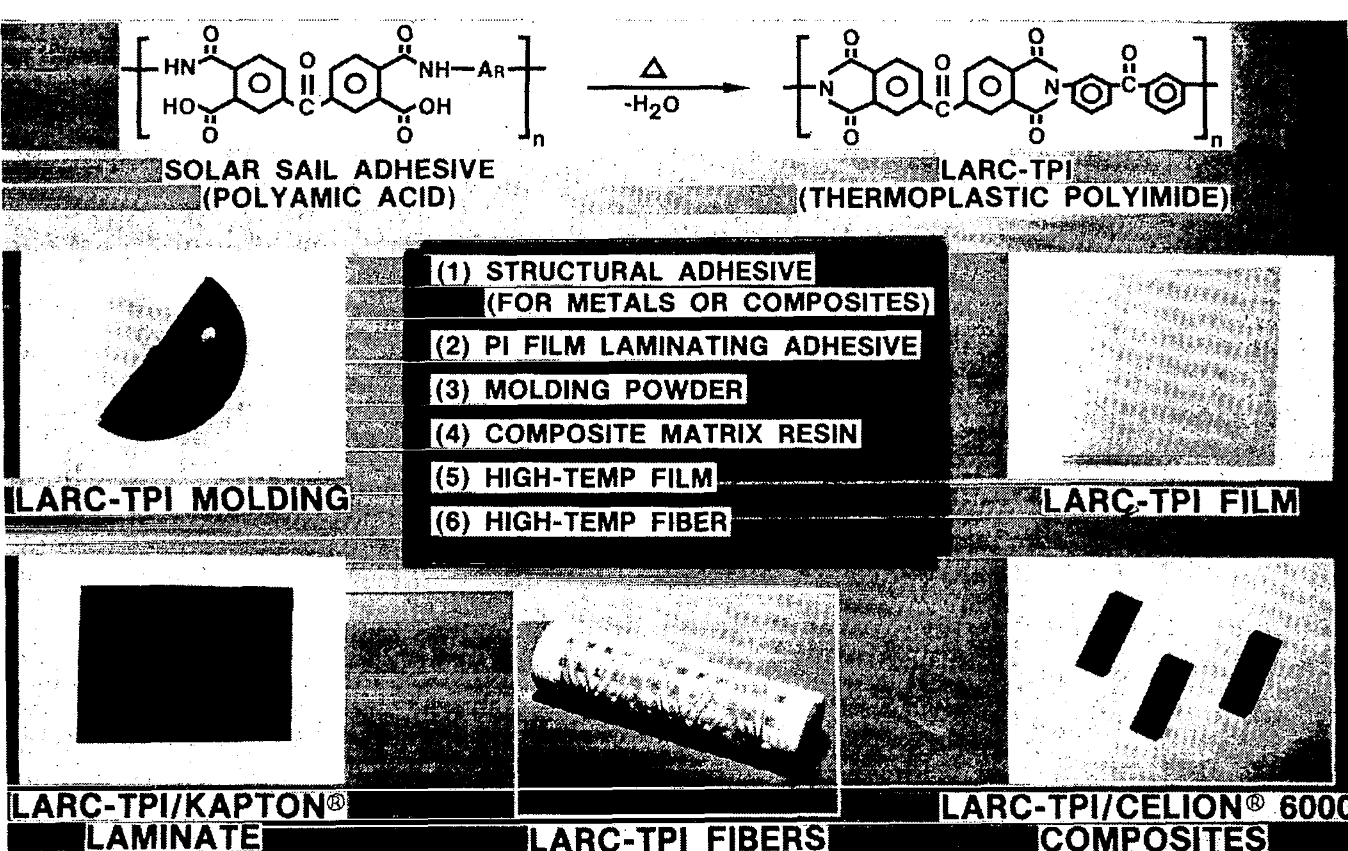
Aromatic polyimides, conversely, are a class of polymers which are known to be resistant to solvents, but they are generally not processable via thermoplastic means. These polyimides are known to be exceptionally thermally stable and like polysulfones and other thermoplastics their use temperature is governed by the softening temperature of each system.

A novel polymer system that possesses the processability of the polysulfones and the solvent resistance of the polyimides has been synthesized and characterized as a film, unfilled molding, filled molding, and adhesivse. The structure of this polyimidesulfone (PISO2) is shown below along with some adhesive and molding data (ref. 8).



LARC-TPI

A linear thermoplastic polyimide, LARC-TPI, has been characterized and developed for a variety of high-temperature applications. In its fully imidized form this new material can be used as an adhesive for bonding metals such as titanium, aluminum, copper, brass, and stainless steel. LARC-TPI is being evaluated as a thermoplastic for bonding large pieces of polyimide film to produce flexible, 100% void-free laminates for flexible circuit applications. The further development of LARC-TPI as a potential molding powder, composite matrix resin, high-temperature film, and fiber will also be discussed (ref. 9).

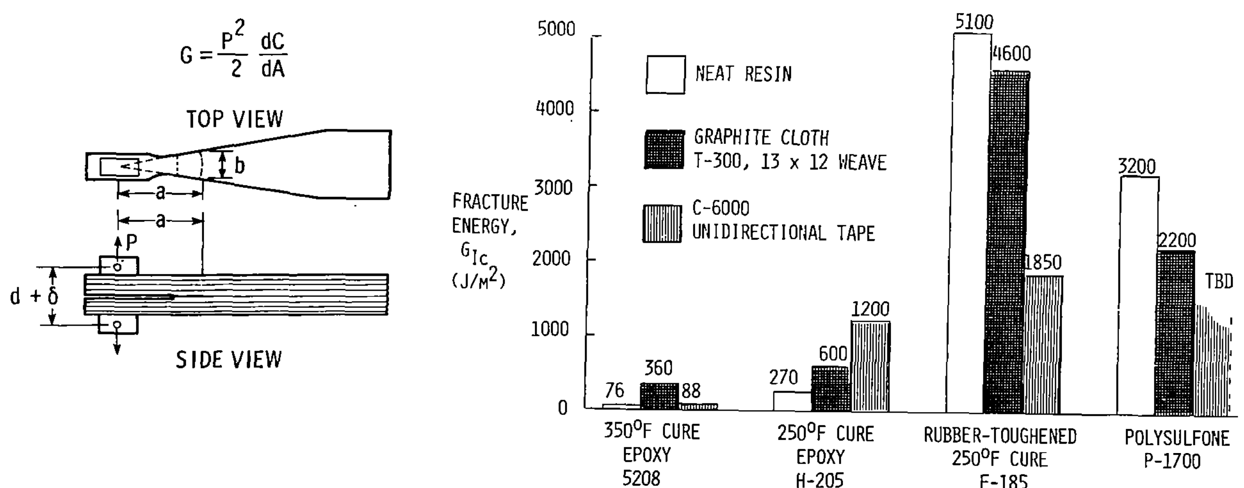


MEASUREMENT OF INTERLAMINAR FRACTURE TOUGHNESS BY COMPOSITE DOUBLE CANTILEVER BEAM TEST

The use of the double-cantilever-beam (DCB) test to measure the fracture toughness (G_{IC}) of adhesives between metal adherends is well known. The application of the test to composite materials in which the crack is initiated in and propagated between two zero plies at the midplane is just now being pursued after the initial work of Bascom et al. (ref. 10). (See left-hand figure below.) Using a width-tapered DCB specimen, these investigators observed a noticeable improvement in fracture toughness of graphite cloth composites made with rubber-toughened epoxies or thermoplastics (P-1700 polysulfone) compared with state-of-the-art 250°F or 350°F cure epoxy systems. (See right-hand figure below.)

An extension of this work to laminates constructed from unidirectional tape (ref. 11 and right-hand figure) indicates that the G_{IC} fracture toughness of the matrix varies widely and unpredictably depending upon the type of reinforcement. This work also shows that the G_{IC} fracture toughness of neat resin cannot be used to predict the G_{IC} value of composites made from that resin. However, the general trend seems to hold that neat resin G_{IC} values are relative indicators of and can help rank fracture toughness in the corresponding composites.

NASA is pursuing an in-depth study of the composite DCB test in which the effects of matrix material, strain rate, specimen dimensions, stacking sequence, and environment are being investigated. A detailed analysis of the failure mechanics of various DCB specimens is also being done to help guide the development of an appropriate pure Mode I interlaminar fracture test method. Key investigators at the National Bureau of Standards, Hercules, Inc., and the University of Illinois, Urbana, are participating in this study.

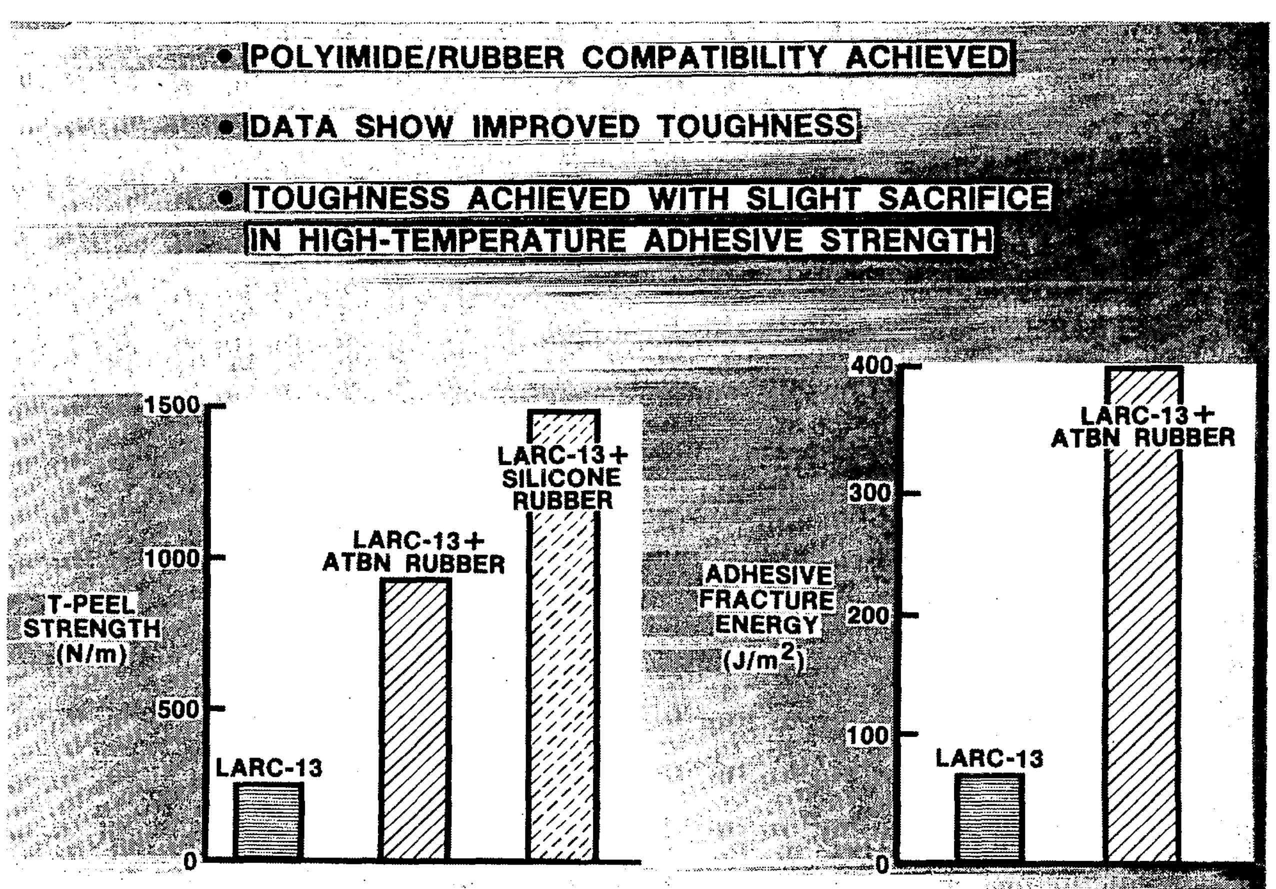


ELASTOMER-TOUGHENED POLYIMIDE ADHESIVES

Addition polyimides are presently being considered as candidate high-temperature adhesives for bonding composite materials and metals such as titanium on future aircraft and spacecraft. These thermoset polyimides undergo cure by an addition reaction involving unsaturated end groups that causes them to be highly crosslinked, insoluble, and extremely brittle.

The elastomer- (rubber) toughening process has been one of the most successful methods for modifying polymer toughness. Incorporation of small amounts of rubber into a polymer matrix has resulted in the significant enhancement of fracture resistance.

This chart illustrates the effects of various added elastomers on the T-peel strength and adhesive fracture energy of a high-temperature addition polyimide, LARC-13 (ref. 12).



MECHANISM OF CURE IN NORBORNENE END-CAPPED IMIDE MODEL COMPOUNDS

Norbornene end-capped imide oligomers such as LARC-160, PMR-15 and LARC-13 display considerable promise for extensive use in various aerospace adhesive and composite applications (ref. 13). These materials were developed in a successful effort to retain the good thermal perfomance of linear condensation polyimides while improving overall processablity. However, very little is known about the mechanism by which these end-capped oligomers cure. This stdy (ref. 14) was designed to increase our fundamental understanding of the fate of the norbornene end-capper as the oligomer is heated.

Model compound I was heated to 285°C in air to yield a partially soluble product mixture that was separated by high-pressure liquid chromatography (HPLC) into three main fractions (A, B, and C). Spectroscopic techniques (NMR, FTIR, MS) were used to prove that the thermal reaction products were geometric isomers. Peak C, the only HPLC peak observed before heating, was starting material and proved to be the kinetically favored endo-endo isomer, II. Peaks B and A were shown to be the endo-exo (III) and exo-exo (IV) configurations, respectively. Further work proved that each isomer thermally isomerized to an equilibrium mixture of all three before further curing reactions took place that rendered the mixture insoluble.

Calorimetry and thermogravimetric analysis indicated that these materials behave differently in air than in nitrogen, suggesting different mechanisms of cure depending upon atmosphere. The data obtained is consistent with a reverse Diels-Alder mechanism leading to loss of cyclopentadiene in nitrogen and a more direct chain extension without weight loss in air.

SAMPLE: I HEATED 285°C/1 hr CH_2Cl_2 EXTRACTED
COLUMN: ZORBAX-SIL
MOBILE PHASE: 1.5% MeOH/ CH_2Cl_2
RATE: 0.7 ml/min
DETECTOR: 254 nm
SIZE: 3μl (1ug/ul)

UV ABSORBANCE
A B C
0 5 10 15
RETENTION TIME, min

NA-p, p'-MDA-NA
I

ENDO-ENDO
II

ENDO-EXO
III

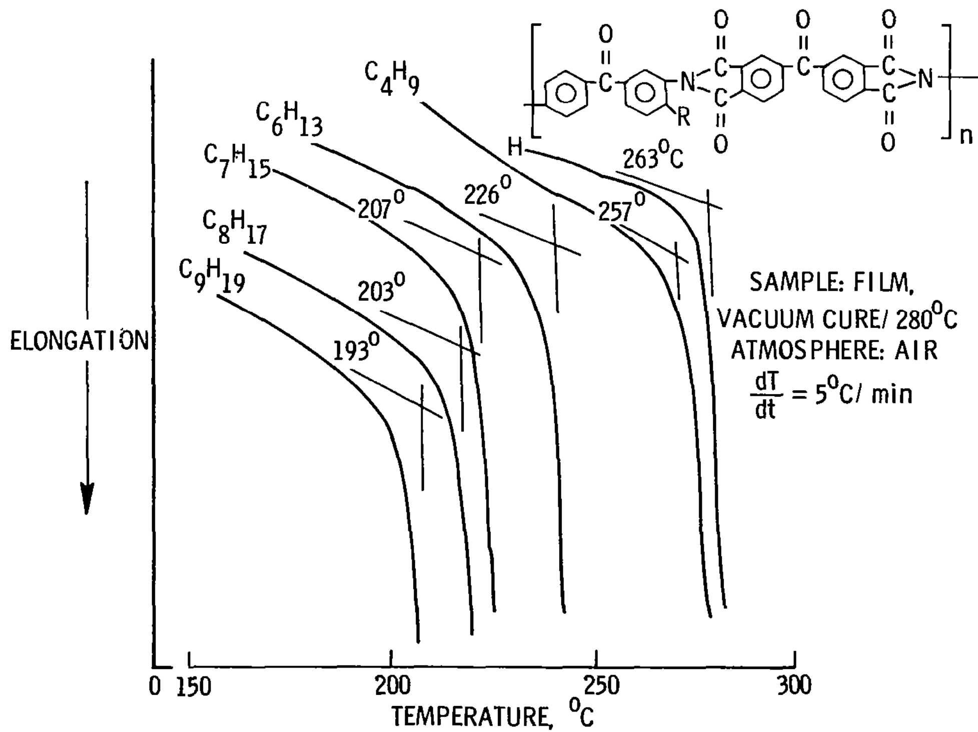
EXO-EXO
IV

EFFECT OF PENDENT ALKYL GROUPS ON POLYIMIDE PROPERTIES

An investigation was conducted to determine the effect on glass transition temperature (T_g), thermal stability and toughness of a polyimide when alkyl groups are attached pendent to the backbone. A series of polymers was prepared in dimethylacetamide (DMAc) using benzophenone tetracarboxylic dianhydride (BTDA) and five different p-alkyl-m,p'-diaminobenzophenones as monomers. The chemical structures are shown in the figure. The alkyl groups varied in length from C_1 (methyl) to C_9 (nonyl).

Poly(amic) acid solutions in DMA_c were vacuum cured to 280°C to afford flexible polyimide films whose T_g decreased with increasing alkyl group length, as determined from thermomechanical analysis. The largest effect, a 70°C decrease in T_g to 193°C, was observed for the polymer containing the nonyl pendent group compared to the T_g of the control polymer (R = H). During thermogravimetric analysis (air, 5°C/min. heating rate), the control exhibited a 10% weight loss at 525°C; the nonyl pendent polymer showed a 10% weight loss at 425°C. The thermooxidative stability of the other films fell between these extremes. Although no increase in the area under the stress-strain curve was observed during film tensile tests, an increase in elongation with a corresponding decrease in tensile strength was noted with increasing alkyl length.

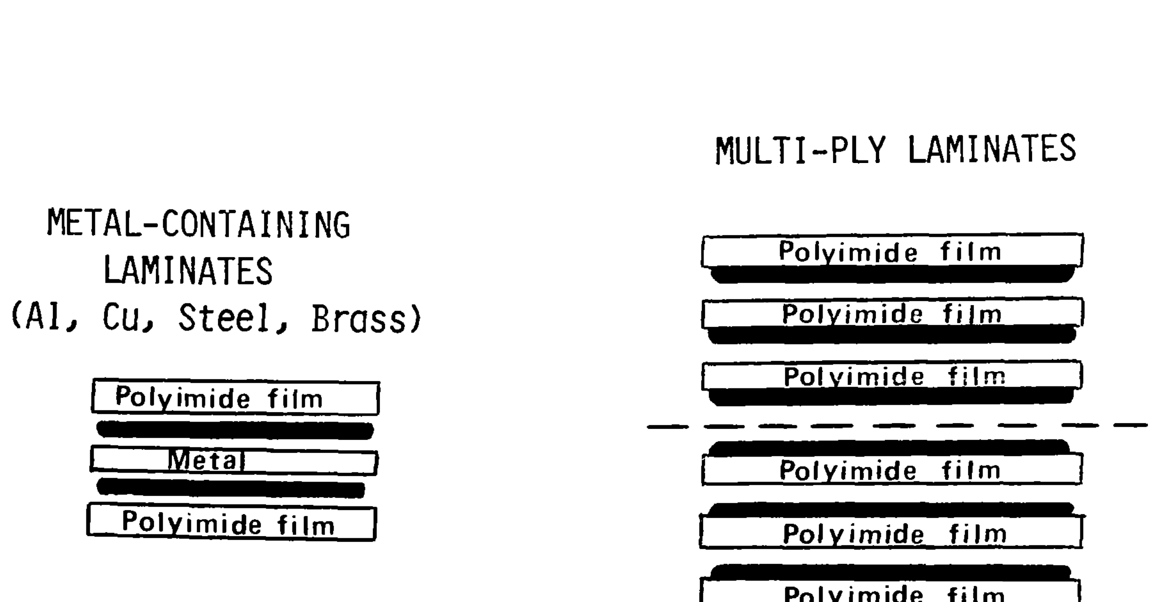
The results of this study indicate that alkyl groups attached pendent to a polyimide backbone can be used to vary the T_g over a wide temperature range. This approach may offer a means of lowering the processing temperature of polyimides without a significant reduction in thermal stability (ref. 15).



FILM LAMINATING

A need exists in the aerospace industry for reliable flexible electrical circuitry that can withstand extreme temperature variations and retain flexibility. Problems to date have been due partially to the presence of voids in film laminates caused by volatiles generated by the adhesive and/or the inherent rigidity of some adhesives. Because it is both flexible and imidized prior to bonding, LARC-TPI shows much potential as a high-temperature adhesive for laminating large areas of polyimide film.

A film-laminating process has been developed whereby films primed with a thin coat of LARC-TPI adhesive are bonded together using temperature and pressure. As an alternate process, LARC-TPI polyamic acid adhesive film may be imidized by heating prior to being sandwiched between polyimide film. When using either process to prouce flexible circuits, a conductive metal may be interposed between layers of the polyimide film. Metal-containing laminates have been made using aluminum, brass, copper and stainless steel sheets or foils (ref. 16).



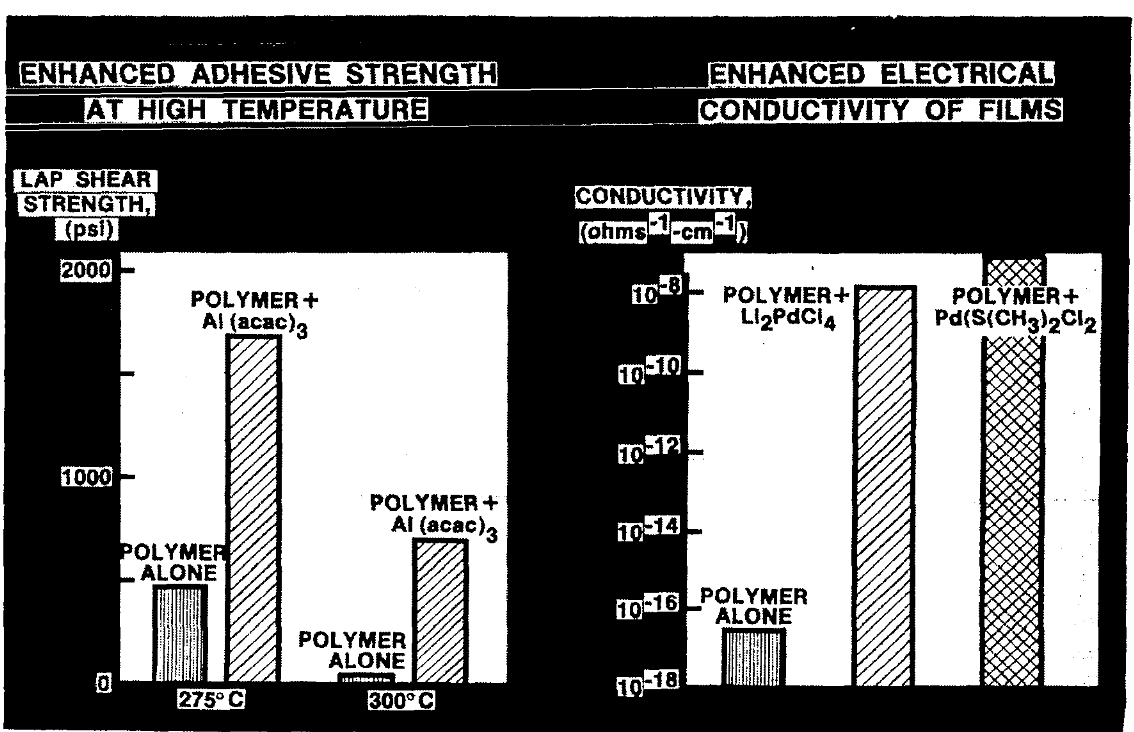
METAL IONS IMPROVE POLYIMIDE PROPERTIES

Polymer films are attractive for various aerospace applications such as antenna surfaces, adhesives, coatings, etc. However, covalently bonded polymers inherently lack the electrical conductivity desirable to resist spacecraft charging or to act as a Faraday Cage and even the most stable polymers developed to date have limited temperature capability.

Several options are available to increase electrical conductivity in a polymer film: (1) mix metallic flakes or powders into the formualtion; (2) laminate metallic/polymer films; and (3) add complex metallic ions to the backbone of the polymer structure. The latter is a very attractive option because the potential for conductivity increases with less increase in the characteristically low polymer film density than is experienced with the other options, but this application has had relatively little attention until recently.

Recent research activity has demonstrated the potential of certain selected metallic ion additions to a polyimide to increase electrical conductivity in a film and high-temperature performance in an adhesive. The addition of the aluminum-ion complex increased adhesive shear strength significantly at 275°C and 300°C. Even more dramatic was the increased electrical conductivity of polyimide films when palladium- and palladium/lithium-ion complexes were added. Electrical conductivity at room temperature was increased by 8 orders of magnitude.

This research is now directed towards elucidation of the mechanisms of electrical (and thermal) conductivity in these metallic ion containing polymers to provide a rational basis for the selection of the most effective ion additions for specific property improvements (ref. 17).



A STUDY OF THE MUTAGENICITY OF AROMATIC DIAMINES

Aromatic diamines are a class of chemicals that are vital for the preparation of high-performance polymers, including the polyurethanes, epoxies, polyamides, and polyimides. However, a number of them are toxic to humans. During the past decade Langley's polymer research has resulted in the synthesis of a broad collection of aromatic diamines with systematic variations in their chemical structures. Although the main reason for acquiring these chemicals was for polymer research, they were also used for a study to learn if any aspects of their chemical structures could be used to predict the mutagenic tendencies of diamines. Mutagenicity is a toxic feature of chemicals that is related to the carcinogenicity of the chemicals.

This comprehensive study has been performed by the Monsanto Research Corporation of Dayton, Ohio on Langley-supplied amines. The investigation disclosed that steric and chemical structural characteristics could be useful in predicting which of the diamines might be mutagenic. The chart below summarizes those results for the steric or spatial isomers (horizontal) of four chemical series (vertical) of aromatic diamines. For example, in general, the electronegative chemical groupings (C = O and especially SO_2) that join the two aniline functions cause the resulting diamines to be less mutagenic (a smaller number or absence of +'s). Conversely, the electropositive coupling groups (CH_2 and especially O) cause the diamines to be more mutagenic. Also, the first three steric isomers on the left tend to be more mutagenic than the three on the right. Unfortunately, many of Langley's accomplishments in polyimide structure-property studies have been achieved using the meta, meta' diamine isomers (third from the left) which are quite mutagenic. But this study also showed that the meta, para' isomers (second from the right) are generally nonmutagenic, so they might be used for polyimides since they give polymers with properties not very different from those made with the meta, meta' diamines.

It is expected that this investigation will extend the usefulness of this novel group of diamine starting materials beyond the original polymer-oriented objectives by providing toxicologists with the means to predict the mutagenic tendencies inherent in aromatic diamines.

Z						
CH_2	+++++	++++	+++	++	–	–
C=O	+++	+++++	++++	–	–	–
SO_2	+	NT	+	–	–	–
O	NT	++++	+++++	++++	+++	NT

CODE: +++++ = VERY STRONG MUTAGEN; ++++ = STRONG; +++ = MODERATE; ++ = LOW; + = VERY LOW; - = NONMUTAGENIC, NT = NOT TESTED

SUMMARY

The polymer program at LaRC involves exploratory studies in polymer science. These include the synthesis of novel polymers and their characterization.

Polymer synthesis programs involve the development of novel thermoplastics, pseudothermoplastics, and thermosets. These systems are prepared to elucidate strucure-property relationships involving thermal capabilities, toughness, processability and environmental stability. Recent investigations have led to the development of more easily processable polyimides, solvent-resistant polysulfones and polyphenylquinoxalines, and tougher high- and intermediate-temperature polymers.

Characterization efforts have included high-pressure liquid chromatography methodology, the development of toughness tests for fiber-reinforced composites, a study of electrical properties of metal-ion-filled polyimides, and a study of the mutagenicity of aromatic diamines. Also the mechanism of cure/degradation of experimental polymers has been studied by rheology, mechanical behavior, separation techniques and spectroscopy. Some of these programs have involved the degradative crosslinking of alkyl-containing polyimides, the separation and identification of crosslinked phenylquinoxalines, the rheological behavior of hot-melt polyimides, and the elucidation of the cure of norbornene endcapped imides.

- o Synthesis Program for Polymers
- o Characterization of Monomers and Polymers
- o Development of Polymers for specific applications
    - Matrix Resins
    - Adhesives
    - Films

REFERENCES

1. St. Clair, T. L.; St. Clair, A. K.; and Smith, E. N.: Structure-Solubility Relationships in Polymers, Academic Press, Chapter 15, pp. 199-215, 1977.

2. Hergenrother, P. M.: Am. Chem. Soc. Div. Org. Coat. and Plastics, 46, 1982, p. 165.

3. Greenwood, T. D.; Armistead, D. M.; Wolfe, J. F.; St. Clair, A. K; St. Clair, T. L.; and Barrick, J. D.: Polymer, 23, Apr. 1982, pp. 621-625.

4. Burks, H. D.; and St. Clair, T. L.: NASA TM-84494, 1982.

5. Hergenrother, P. M.: Macromolecules, 14, 1981. pp. 891, 898.

6. Hergenrother, P. M.: Polymer Eng. and Sci., 21 (16), 1981, p. 1072.

7. St. Clair, A. K.; and St. Clair, T. L.: Polymer Eng. and Sci., 22 (1), 1982, pp. 9-14.

8. St. Clair, T. L.; and Yamaki, D. A.: A Thermoplastic Polyimidesulfone, to be presented at First Technical Conference on Polyimides, Society of Plastics Engineers, Inc. (Ellenville, NY), Nov. 10-12, 1982.

9. St. Clair, A. K.; and St. Clair, T. L.; SAMPE Quarterly, 13 (1), 1981, pp. 20-25.

10. Bascom, W. D.; Bitner, J. L.; Moulton, R. J.; and Siebert, A. R.: Composites, Jan. 1980, p. 9.

11. O'Brien, T. K.; Johnston, N. J.; Morris, D. H.; and Simonds, R. A.: SAMPE Journal, 18 (4), 1982.

12. St. Clair, A. K.; and St. Clair, T. L.: International J. of Adhesion and Adhesives, 1 (5), 1981, pp. 249-255.

13. Dexter, H. B.; and Davis, J. G. (Eds.): Graphite/Polyimide Composites, NASA CP-2079, 1979.

14. Young, P. R.: NASA TM-83192, 1981.

15. Jensen, B. J.; and Young, P. R.: Polyimide Characterization Studies: Effect of Pendent Alkyl Groups, to be presented at First Technical Conference on Polyimides, Society of Plastics Engineers, Inc. (Ellenville, NY), Nov. 10-12, 1982

16. St. Clair, A. K.; and St. Clair, T. L.: U.S. Patent Application, serial no. 189,234, Sept. 22, 1980.

17. Taylor, L. T.; St. Clair, A. K.; Carver, V. C.; and Furtsch, T. A.: U.S. Patent 4,311,615, Jan. 19, 1982.