Structure-property relations in processing high-performance polyisoimide–imide resins

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(Received 20 December 1991; accepted 15 May 1992)

Synopsis

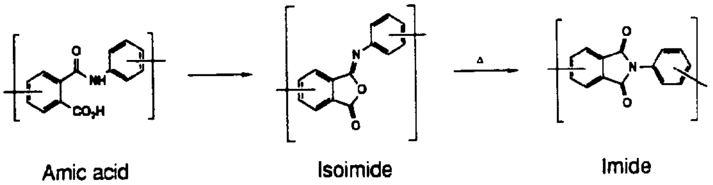
The processing of high-performance polyimides via their processible isoimide modifications is of great interest to aeronautical and aerospace applications. Rheological characterization during the isoimide-to-imide conversion, essential for identifying the processing conditions of such reactive polymers and their polymer blends, is reported. Using dynamic shear measurements, we obtained rheological information for a series of polyisoimide–imide resins as a function of temperature and cure history. Both homopolymers and copolymers have been investigated. We found that the processibility (e.g., melt viscosity of isoimide) and ultimate product properties (e.g., T_g of polyimide) can be systematically varied by changing the chemical constituents and rigidity of the polymer backbone. Such correlations of chemical structures, rheological behavior, and processing characteristics provide the necessary database for tailor making new polymer systems with desirable processibility and mechanical properties. Practical considerations and difficulties in measuring the viscoelasticity of these materials are also addressed.

INTRODUCTION

Development of lightweight polymer composite structural components for future aerospace vehicles is important for civil, military, and space transport systems, particularly in supersonic transport. Materials technology must satisfy the requirements of cost effectiveness and durability, while maintaining high toughness and tensile modulus at temperatures well beyond 370 °C (700 °F), and achieving environmental and high-energy radiation resistance.

Polyimides are potentially suitable because of their demonstrated ultrahigh temperature tolerance and well-established chemistry. They are capable of retaining useful mechanical properties even after exposure to chemicals, irradiation, and thermal, atmospheric, electrical, and mechanical stresses over time (Mittal, 1984). Thermoset polyimides, the bismaleimides, are relatively easy to process but they lack the toughness required for some structural applications. Thermoplastic polyimides, on the other hand, have a high level of fracture toughness but are difficult to process.

Isoimide modification has been shown to be a viable and attractive approach to achieve enhancement in processibility of polyimides (Kreuz, 1968; Awad *et al.*, 1977; Lau, 1983; Landis and Lau, 1990). The chemistry of the isoimide conversion is illustrated as follows:



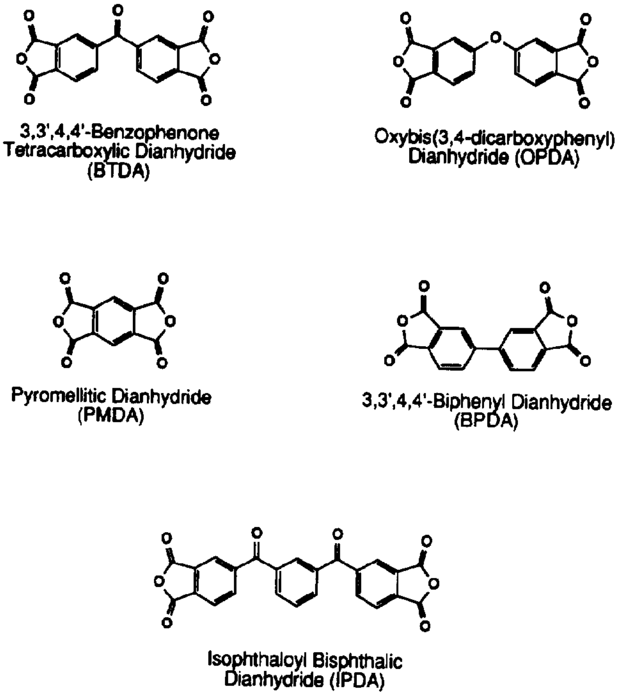
Isoimide is the processible isomeric form of the imide. Its asymmetry generally contributes to higher solubility and lower melt viscosity than the imide form. The isoimide functionalities convert to the imide form without evolution of volatiles, resulting in minimal volume change and void formation. The chemical transformation from monomers to the polyamic acid, which in turn forms the isoimide and imide, is well-defined chemistry and provides reproducible synthesis and thus easy quality control.

We report herein the results from our study on the rheological behavior of the isoimide-to-imide conversion. One objective of this work is to develop characterization methods that can be used to define processing windows and procedures for the polymer resins and to assess the relative processibility. The polymer backbone structure has been varied systematically to develop structure/property/processing relationships needed for tailoring material properties for specific applications.

EXPERIMENTAL

Polymer synthesis and characterization

The general synthetic technique for the preparation of various polyisoimides of different molecular weights entails: (1) the formation of the polyamic acid precursor by the reaction of stoichiometric amounts of acid dianhydride, diamine, and end-capping agent phthalic anhydride



3,3',4,4'-Benzophenone Tetracarboxylic Dianhydride (BTDA)

Oxybis(3,4-dicarboxyphenyl) Dianhydride (OPDA)

Pyromellitic Dianhydride (PMDA)

3,3',4,4'-Biphenyl Dianhydride (BPDA)

Isophthaloyl Bisphthalic Dianhydride (IPDA)

FIG. 1. Chemical structures of aromatic dianhydrides for polyimide and polyisoimide synthesis.

(PA), in an appropriate solvent such as tetrahydrofuran (THF), *N*,*N*-dimethylacetamide (DMAC), and *N*-methylpyrrolidinone (NMP); (2) cyclodehydration of the polyamic acid with a stoichiometric quantity of *N*,*N*′-dicyclohexylcarbodiimide (DCC) to yield the polyisoimide and the insoluble by-product, *N*,*N*′-dicyclohexylurea (DCU), which can be removed by filtration; and (3) precipitation of the polyisoimide from the reaction solution with isopropyl alcohol or acetone. Figures 1 and 2 illustrate the chemical structures of the aromatic dianhydrides and diamines, respectively, used for the polymer synthesis. More detailed synthesis procedures and reaction parameters on specific polyisoimides can be found in Landis *et al.* (1991a and 1991b).

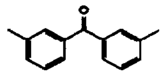
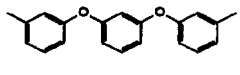
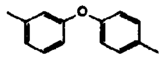
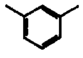
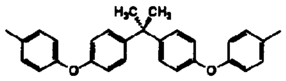
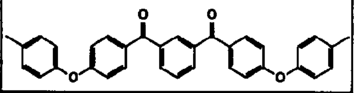
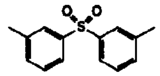
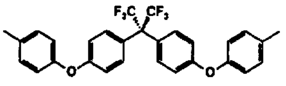
Diamines H_2N-Ar-NH_2		
Name	**Acronym**	**Ar**
3,3'-Diaminobenzophenone	DAB	
1,3-Bis(3-aminophenoxy)-benzene	APB	
3,4'-Oxydianiline	3,4'-ODA	
1,3-Diaminobenzene (*meta*-Phenylenediamine)	MPDA	
2,2-Bis[4-(4-aminophenoxy)-phenyl]propane	BAPP	
1,3'-Bis[4-(4-aminophenoxy)-benzoyl]benzene	1,3'-BABB	
3,3'-Diaminodiphenylsulfone	3,3'-DDS	
2,2-Bis[4-(4-aminophenoxy)-phenyl]hexafluoropropane	BDAF	

FIG. 2. Chemical structures of aromatic diamines for polyimide and polyisoimide synthesis.

Fourier-transform infrared spectroscopy (FTIR) studies generally provide qualitative information on the batch-to-batch variance in synthesis and the conversion of isoimide to imide. Figure 3 shows comparative FTIR spectra of a representative polyisoimide, poly(BTDA-APB) isoimide, before and after thermal isoimide-to-imide conversion. The characteristic infrared absorptions of polyisoimides are the carbonyl band at 1790–1841 cm^{-1} and the imine band at 1680–1730 cm^{-1}, both

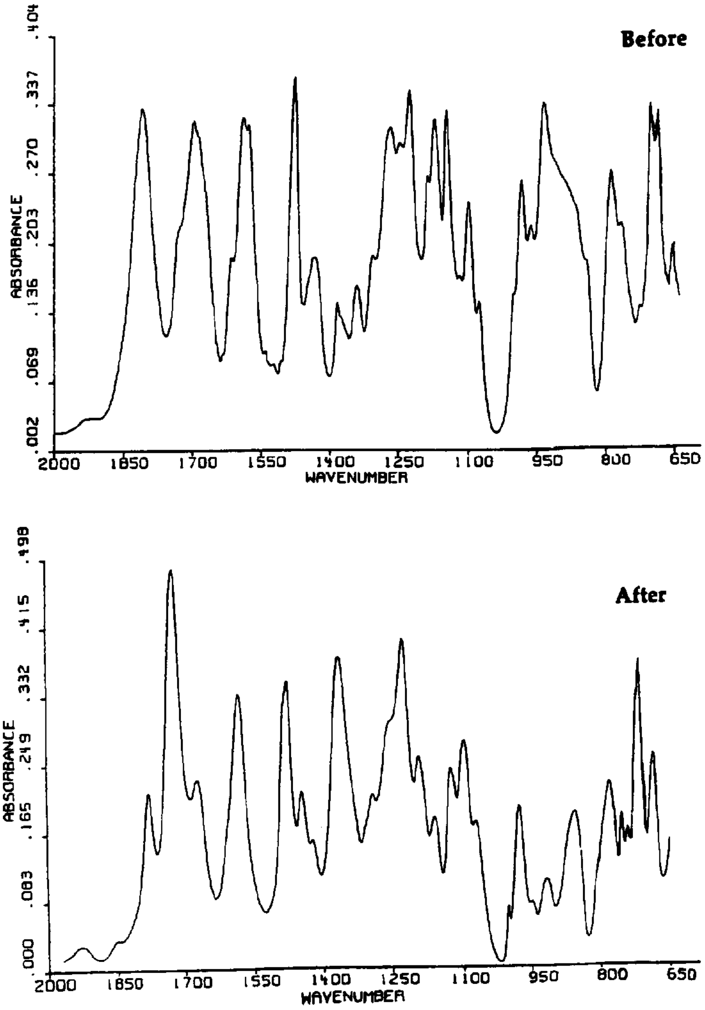


FIG. 3. Fourier-transform infrared spectra of poly(BTDA-APB)isoimide before and after thermal imide-to-isoimide conversion.

intense. The characteristic broad band at 900–950 cm^{-1} is attributable to the lactone ring with an exocyclic double bond. The isoimide-to-imide conversion can be routinely monitored by the disappearance of these bands with the concomitant growth of the characteristic polyimide absorptions at 1776–1795 (asymmetric), 1707–1730 (symmetric), and at 1380 cm^{-1}. FTIR measurements was performed on a Nicolet model 60SX spectrometer.

All thermochemical behavior and glass transition temperature (T_g) of the polyisoimides were obtained by differential scanning calorimetry (DSC). DSC specimens after the temperature ramp were cooled down slowly to ambient and rescanned to 400 °C to capture the T_g of the corresponding polyimides. Data were obtained from 10 to 15 mg samples in a nitrogen atmosphere at a 10 °C/min heating rate using a Perkin-Elmer DSC-7 analyzer.

Rheological characterization

Specimens suitable for rheological measurements were prepared from the powdery polyisoimides by pressing circular pellets from a vacuum compression mold, using a hydraulic press at ambient temperature. A predetermined quantity of polyisoimide powder was first placed inside the mold cavity, and the cavity was connected to a vacuum pump at 10^{-3} mm pressure for 10 min to remove trapped air in the polymer sample. Then a pressure of 12 000 psi was applied to the plunger which compresses the powder into a pellet of 25, 8, or 5 mm in diameter and 2 mm in thickness. Prior to forming a pellet, the powdery polymer was first pretreated at 175 °C *in vacuo* for 30 min to remove residual solvent or moisture.

The flow properties of polyisoimides as a function of temperature were obtained using a Rheometrics mechanical spectrometer, RMS-800, which is equipped with an environmental chamber for temperature control. Nitrogen purge was used during all runs to avoid oxidation of the samples. Oscillatory shear (dynamic shear) mode and parallel plates were used to measure the dynamic viscosity at a frequency of 10 rad/s. A temperature sweep program was used to control the change in temperature.

After the specimen was placed between the parallel plates at ambient temperature, the temperature of the environmental chamber was allowed to equilibrate at 140 °C (below the polymer melting temperature). The temperature was programmed to rise to 190 °C at 10 °C per increment, to 300 °C at 2 or 5 °C per increment, and then to 400 °C at 10 °C per increment. (In some cases, the program was terminated earlier due to sample outgassing.) The dynamic viscosity (η^*) was measured after the chamber temperature stabilizes at each temperature step. The actual temperature of the specimen was monitored by a thermocouple in contact with the platen on which the specimen was placed. The specimen was subjected to a slight compression during the experiment to minimize slippage between the material and the platens. The percent strain applied to the specimen was varied using the autostrain option of the program to control the deformation so that it was large enough to produce a measurable stress signal during the entire temperature scan.

Dynamic properties of thermally converted polyimides as a function of temperature were obtained by immediately rescanning cooled samples that had been subjected to one rheometric temperature cycle. Adhesion of samples to the platens in this case was excellent due to melting of the polyisoimide in the first cycle.

RESULTS AND DISCUSSION

Homopolymers

Figure 4(a) shows the rheological response of poly(BTDA-DAB) isoimide in comparison with its corresponding imide (LARC-TPI) as a function of temperature. This rheological profile is characteristic of this class of polymers. At temperatures below 190 °C (region a), before the specimen softens, there is poor adhesion between the sample and the parallel plates fixture, leading to a significant amount of slippage at the solid boundaries. Therefore, the results are not meaningful in this region. As the polyisoimide is heated to over 190 °C (region b), the softening of polyisoimide improves adhesion to the fixtures and wall slip diminishes. The measured dynamic viscosity decreases as the polymer softens or melts more with increasing temperature. Concurrently, the isoimide undergoes a thermally induced chemical conversion to the corresponding imide isomer at these temperatures (region c), causing an increase in η^*. These two competing mechanisms result in a minimum in the viscosity profile (η_{min}) to occur at some intermediate temperature (T_{min}), about 243 °C for poly(BTDA-DAB)isoimide, above which the effects of the chemical conversion begin to dominate (region d). Further increase in temperature completes the isoimide-to-imide conversion (region e). Upon a second temperature scan of the imidized polymer, the viscosity minimum disappears [see Fig. 4(b)]. Instead, the polymer exhibits high-performance properties, showing high dynamic storage modulus ($G' > 10^9$ dyn/cm^2) at low temperatures and a T_g of 260 °C.

The rheological profile obtained for the isoimide-to-imide conversion can be used to determine processing conditions. The viscosity minimum

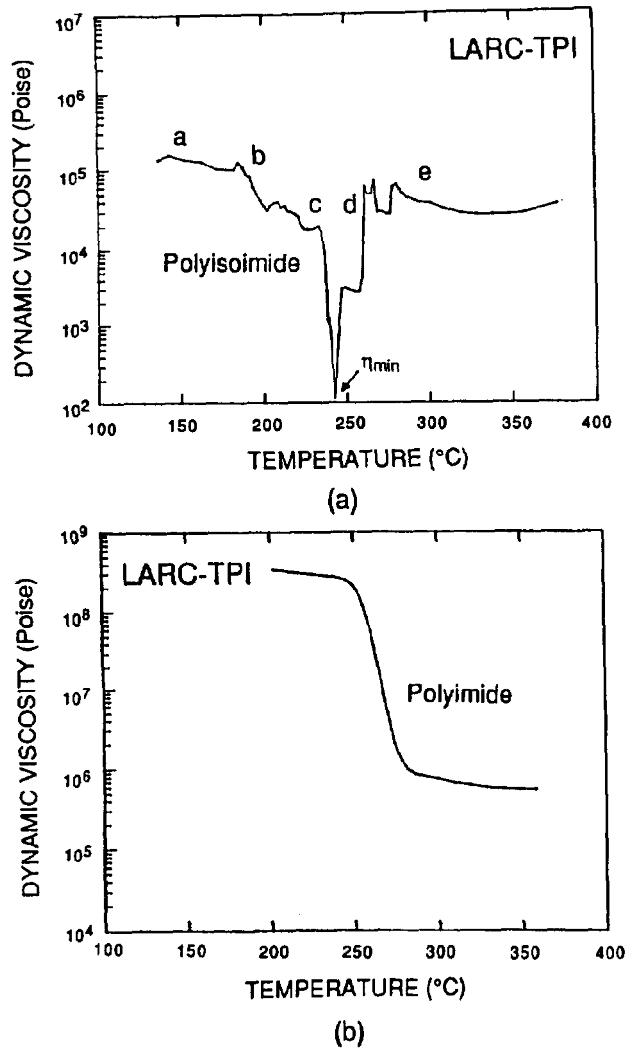


FIG. 4. Dynamic viscosity as a function of temperature (a) during conversion from poly(BTDA-DAB)isoimide to its corresponding polyimide (LARC-TPI), and (b) after complete conversion to polyimide.

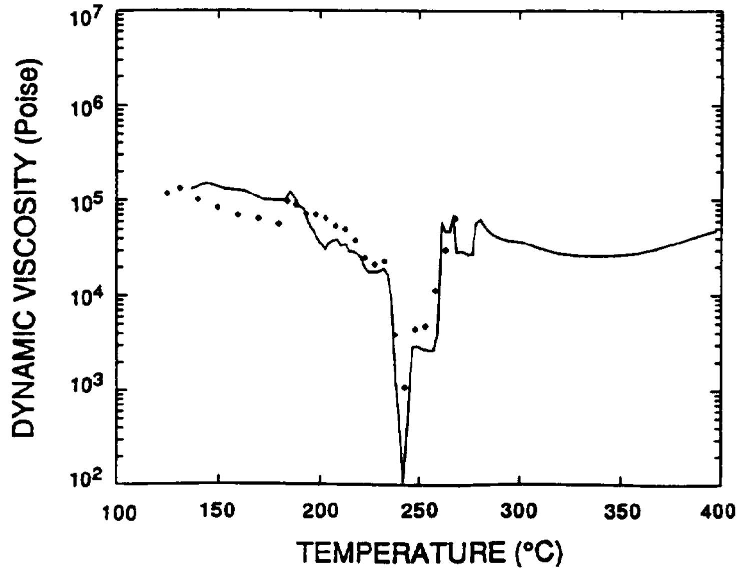


FIG. 5. Dynamic viscosity as a function of temperature of poly(BTDA-DAB)isoimide at two different scanning rates: solid line, 2 °C per increment; diamonds, 5 °C per increment.

η_{min}, as discussed above, results from a competition between melting of polyisoimide (lower η^* with increasing temperature) and conversion to polyimide (higher η^* with increasing temperature). Although it is not a fundamental quantity and is dependent on the measurement procedures, we found that η_{min} can be used as an empirical tool to rank the processibility of polyisoimides, as the scanning rates chosen (an average of 1.6 and 3.5 °C/min for the 2 and 5 °C increments, respectively) are comparable to those in actual processing. As illustrated in Fig. 5, both the 2 and 5 °C increment scans from 190 to 300 °C yield essentially identical results within the experimental uncertainty in determining T_{min}. The value of η_{min}, however, can be determined more accurately from the 2 °C increment scan since 5 °C is sometimes too large of an increment to catch the minimum in the profile, especially when the drop in the dynamic viscosity is very sharp as in the case of poly(BTDA-DAB)isoimide. T_{min} defines the processing window for prepregging and/or consolidation if these materials are to be used as resin matrix for composites. The rheological data demonstrate the main advantage of

TABLE I. Comparison of processibility between LARC-TPI and its isoimide analog.

Poly(BTDA-DAB)imide (LARC-TPI)	Poly(BTDA-DAB)isoimide (isoimide analog)
Insoluble (DMAC)	Soluble (DMAC, amides)
Processed by powder method	Facile film forming
Prepregged from amic acid	Potential for volatile-free prepegging
Volatiles during processing	Easy isoimide-to-imide conversion without volatiles
High melt viscosity	Lower melt viscosity and processing temperature

the isoimide route to polyimide: the resin is rendered more processible at low temperatures (near the viscosity minimum) but retains the high T_g needed under use conditions.

The isoimide modification technique generally improves not only melt but also solution processibility of imide-containing polymers; previously unprocessible linear polyimides can now be used in polymer blending. Table I, a comparison between LARC-TPI polyimide and its isoimide analog, illustrates an example of enhanced processibility. Our preliminary studies showed that complete miscibility was achieved over a wider variation of the ratio of the thermoplastic and thermoset components. The final product is indistinguishable from the composition that would be obtained if it were possible to start with the imide forms of the components.

Ideally, the rheological profile most practical for processing is one with a low value of $\eta_{\min}$ and a wide window near $T_{\min}$ with the low viscosity. Figure 4 is an example of a profile with a sharp drop in the viscosity and thus a relatively narrow processing window. Figure 6 illustrates a different profile, for poly(BTDA-APB)isoimide, which is characterized with a wider "valley" around $T_{\min}$ but higher value of $\eta_{\min}$.

In these experiments, the observed rheological profile is a function of the temperature scanning rate, the melting rate of polyisoimide, and the reaction kinetics of the isoimide-to-imide conversion. Nevertheless, the use of the rheological characterization procedures developed here to rank processibility can be justified since our method for probing a chemical transition is analogous to the widely accepted measurements of the glass transition temperature (T_g) using differential scanning calorime-

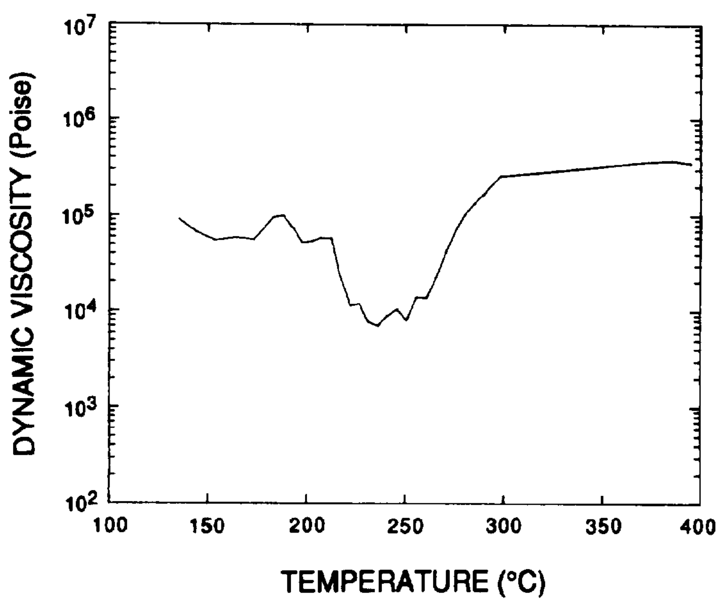


FIG. 6. Dynamic viscosity as a function of temperature during conversion from poly(BTDA-APB)isoimide to its corresponding polyimide.

try (DSC) for probing a physical transition. In both cases, the parameters of interest (T_{min}, η_{min}, and T_g) are scan-rate dependent, but a judicious selection of scanning parameters allows us to utilize the measured quantities in a meaningful (albeit empirical) manner to rank materials and their usefulness in applications. Furthermore, as in the case of T_g measurements, T_{min} and η_{min} have been found to be relatively insensitive to small variations in scan rate (see Fig. 5), thereby demonstrating the robustness of the characterization procedures for these polymers. We speculate that such insensitivity of the profile to scan rate is because both rates chosen in this study are fast compared to the conversion reaction at temperatures below T_{min} as indicated by very limited kinetic data obtained in our laboratory using FTIR to monitor the disappearance of the isoimide peaks. Consequently, regions b and c of Fig. 4 reflect mostly the melting of polyisoimide. As in the case of T_g measurements, one does not need a detailed knowledge of the kinetics of the transition to make use of the rate-dependent physical measurements

TABLE II. Magnitude of changes in dynamic viscosity from polyisoimide to polyimide.

State	η^*(P) (at 10 rad/s)
Polyisoimide below T_m	10^9
Polyisoimide at T_{min}	10^2–10^4
Polyimide above T_g	10^6–10^7
Polyimide below T_g	10^9

made in the vicinity of the transition. Moreover, a quantitative measure of the detailed reaction kinetics of such complex systems is out of the scope of our current work.

Because of the large variations (several orders of magnitude) in the rheological functions during the isoimide–imide conversion as illustrated in Table II, dynamic shear is the most appropriate characterization method for assessing the rheology of these materials. However, due to the high modulus of the polyimides, transducer compliance could be a problem especially for the relatively large sample size of 25 mm in diameter. Thus, to obtain an accurate temperature dependence behavior for the polyimide, the pellet size has to be reduced to 8 to 5 mm in diameter. On the other hand, such small samples often do not provide a large enough stress signal near η_{min}; a larger sample is then needed if accurate values near the minimum viscosity during the conversion is desired. After completion of the conversion to polyimide (region e in Fig. 4), the transducer compliance could again be a problem for the high modulus material, rendering large errors in the η^* measurements in that region. Finally, the slight normal force (less than 100 g), which has been applied to the pellet initially to enhance adhesion to the parallel plates fixture as the polyisoimide softens, may also affect the accuracy of the measured dynamic viscosity values. Nonetheless, with consistent sample loading and measurement procedures, we found that the reproducibility of data is usually less than 30% variations in η_{min} and ± 3 °C for T_{min}. These variations arise from the complexity of the dynamics and the difficulties in obtaining meaningful rheological measurements for these materials; but they are acceptable for our purposes.

Correlation with molecular weight

The value of the η_{min} exhibited by polyisoimides upon increasing temperature was found to depend on the polymer molecular weight. For the same polymer backbone structure, the lower molecular weight poly-

TABLE III. Correlation of viscosity minima with degree of polymerization of polyisoimides.

Polymer constituent				
Dianhydride	Diamine	DP[a]	$\eta_{\min}$ ($\times 10^{-4}$ P)	$T_{\min}$ (°C)
BTDA	BAPP	50	4.16	213
		10	1.65	235
PMDA	APB	50	8.10	257
		10	6.19	171
BTDA	3,3′-DDS	50	4.72	241
		10	2.76	218
BPDA	APB	50	2.92	196
		10	0.440	208
OPDA	APB	50	0.451	197
		10	0.0323	194

[a] DP = degree of polymerization.

isoimide generally shows a lower value for the minimum than its higher molecular weight analog as shown in Table III. $T_{\min}$, on the other hand, does not seem to be correlatable to the molecular weight.

Correlation with chemical structure

Similar to the variation of T_g values with chemical structures, the more rigid structures tend to impart higher melt viscosities to the polyisoimides. Within a series of polyisoimides based on a common diamine (e.g., APB) and the same degree of polymerization (DP = 50), melt viscosity increases in the order of increasing rigidity of the dianhydride, OPDA < BTDA < BPDA < PMDA, as shown in Fig. 7(a). Similarly, in the series of polyisoimides based on the dianhydride BTDA, e.g., APB < 3,3′DDS < MPDA or 3,4′-ODA < 3,3′DDS < MPDA, melt viscosity increases in the order of increasing rigidity in the diamine [Fig. 7(b)].

It is interesting to note that the apparent added flexibility in the extended chain length and an extra oxy-linkage in APB, as compared to 3,4′-ODA, does not result in a lower value for the minimum melt viscosity of the polymer poly(BTDA-APB)isoimide. This result is consistent with a similar observation that the apparent flexibility in the extended chain length and oxy-linkages in the diamine components of poly(BTDA-1,3′-BABB)isoimide and poly(BTDA-BAPP)isoimide does not result in lower melt viscosities. The high melt viscosities are probably due to the increase in molecular weight within the same degree of polymerization.

POLYISOIMIDE SERIES BASED ON DIAMINE APB	OPDA		BTDA		BPDA		PMDA	
	η^*	T_{min}	η^*	T_{min}	η^*	T_{min}	η^*	T_{min}
	4510	197	7020	236	29200	196	81000	257

(a)

POLYISOIMIDE SERIES BASED ON DIANHYDRIDE BTDA	APB		3,4'-ODA		3,3'-DDS		MPDA	
	η^*	T_{min}	η^*	T_{min}	η^*	T_{min}	η^*	T_{min}
	7020	236	2880	243	47200	241	73000	234

(b)

FIG. 7. Correlation of viscosity minima of polyisoimides with structural variation of (a) dianhydrides and (b) diamines.

Copolymers

Preliminary small laminate data from our laboratory indicated that copolymers derived from LA-100 [poly(BTDA-APB)isoimide] and poly(BTDA-3,4'-ODA)isoimide showed good retention of hot/wet properties at 350 °F. Several phthalic anhydride-end-capped polyisoimides and copolyisoimides based on BTDA, APB, and 3,4'-ODA were thus synthesized. Figure 8 summarizes some structure-property correlation data for this series of copolyisoimides.

The T_g correlation suggests that the introduction of the more flexible component APB into poly(BTDA-3,4'-ODA)imide lowers the T_g of the material. The relationship between polymer structure and melt viscosity appears to be more complicated, the melt viscosity of the 1:1 copolymer having a higher value than those of the 1:3 and the 3:1 copolymers or of the homopolymers.

Figure 9 summarizes some structure-property correlation data for the BDAF-based series of copolyisoimides. The dianhydride component of these polyisoimides comprises different molar ratios of the dianhydrides BTDA and the more rigid PMDA. The T_g correlation suggests that the introduction of the more rigid dianhydride PMDA into

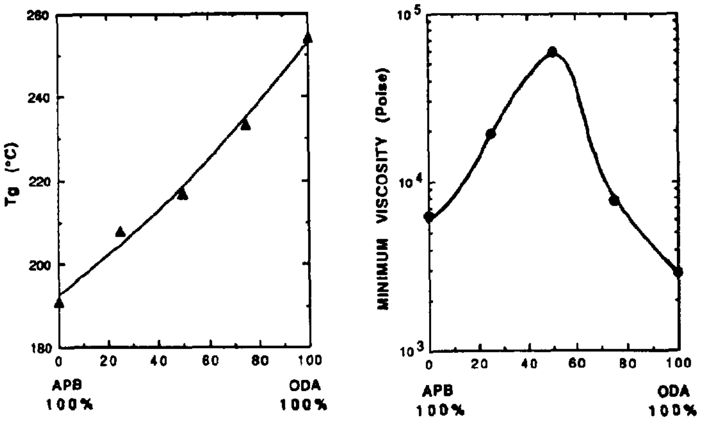


FIG. 8. Glass transition temperature and viscosity minimum correlation of copolyisoimides of dianhydride BTDA and diamine mixtures of APB and 3,4′-ODA.

poly(BTDA-BDAF)imide raises the T_g of the material. The relationship between polymer structure and melt viscosity appears to be similar to the copolymers of poly(BTDA-APB)isoimide and poly(BTDA-3,4′-ODA)isoimide discussed above. The melt viscosity of the 1:1 copolymer has a higher value than those of the 70:30 and the 30:70 copolymers or of the homopolymers. The range of variation, however, is not as large as the copolymer series of poly(BTDA-APB)isoimide and poly(BTDA-3,4′-ODA)isoimide.

Effects of temperature history on processibility

In composite manufacturing, processing conditions used in one step (e.g., prepregging) often have a significant impact on the processibility in subsequent steps (e.g., consolidation) and on product properties. For any reactive systems such as isoimide polymers and blends, the temperature history seen by the materials plays a particularly important role in their processibility. In a series of experiments on poly(MPDA-IPDA)isoimide, we included a lengthy isothermal hold at various temperature below T_{min} in order to study the effects of temperature history on the rheological profile. Figure 10(a) shows the profile of a control experiment using the established temperature scan procedure with no isothermal hold. This polymer exhibits a T_{min} of about 252 °C. Figures 10(b)–10(d) illustrate the rheological profiles as a function of temper-

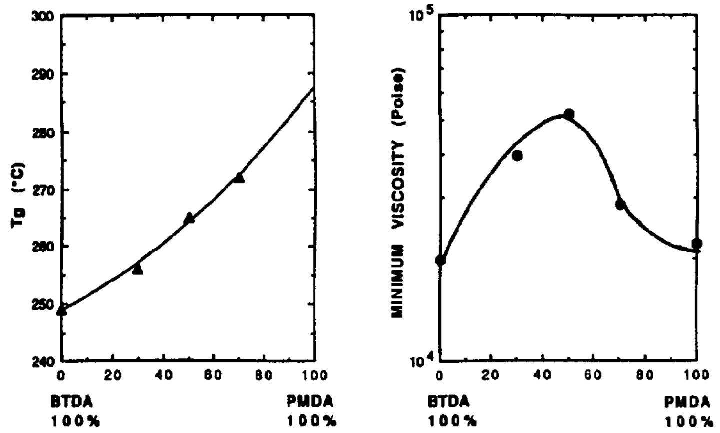


FIG. 9. Glass transition temperature and viscosity minimum correlation of copolyisoimides of diamine BDAF and dianhydride mixtures of BTDA and PMDA.

ature with an added temperature hold of 60 min at 200, 240, and 250 °C. As expected, the higher the isothermal preconditioning temperature, the bigger the impact on the value of η_{min} and thus processibility. Interestingly, whereas η_{min} increases by up to an order of magnitude, T_{min} remains relatively constant. This trend supports our hypothesis that the profile prior to T_{min} reflects the melting of polyisoimide. As some isoimide functionalities are converted to imide during the isothermal preconditioning, the relative values of η_{min} within one polymer system reflect the portion of isoimide remaining. From a practical standpoint, our rheological data suggest that the processing window defined empirically by T_{min} is fairly insensitive to the temperature history seen by the material.

CONCLUSIONS

Our study illustrates that the isoimide modification approach to enhance the processibility of imide-containing high-performance polymers can be quantified using rheological characterization of the reactive isoimide polymers as a function of temperature. The rheological profiles measured using practical scan rates allow us to empirically rank processibility and define processing windows for these materials. We have investigated the structure/property relationships of homopolymers and

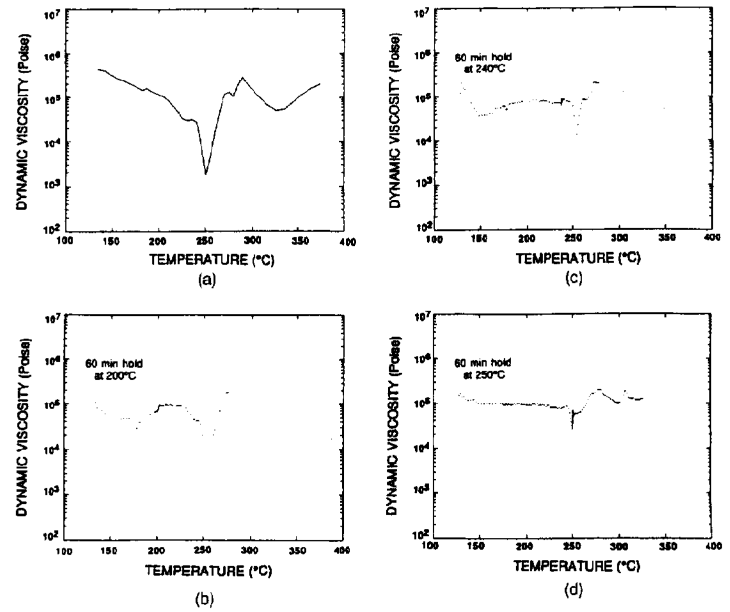


FIG. 10. Dynamic viscosity as a function of temperature during conversion from poly(MPDA-IPDA)isoimide to its corresponding polyimide with (a) no isothermal hold, (b) an isothermal hold of 60 min at 200 °C, (c) an isothermal hold of 60 min at 240 °C, and (d) an isothermal hold of 60 min at 250 °C.

copolymers. Due to the increased solubility of polyisoimides, blends of novel combinations of thermoplastic and thermoset polyimides can now be explored and may result in economical, tailored-made engineering polymers for aerospace and commercial applications (Pascal *et al.*, 1989; Turpin and Green, 1990; Pater, 1990). The characterization procedures developed will be useful to study the rheology and processing of blends containing polyisoimides.

ACKNOWLEDGMENTS

This work is supported by the NASA Program "Advanced Composites Structural Concepts and Materials Technologies for Primary Aircraft Structures," NAS1-18888. The authors are indebted to NASA-Langley and Lockheed Aeronautical Systems Company for financial support. They wish to thank Mr. Anthony C. Jackson (Lockheed Aeronautical Systems Company), Dr. Paul M. Hergenrother, Dr. Norman J. Johnston, and Dr. Terry L. St. Clair (Langley Research Center) for their encouragement and guidance. The authors also wish to express their gratitude to Ms. Martha M. Steiner and Mr. Anthony L. Green for thermal analyses, and to Ms. Sandra K. Clement for synthesis support.

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