Low Viscosity Imides Based on Asymmetric Oxydiphthalic Anhydride

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A series of low-melt viscosity imide resins were prepared from asymmetric oxydiphthalic dianhydride (a-ODPA) and 4-phenylethynylphthalic anhydride as the endcap, along with 3,4′-oxydianiline (3,4′-ODA), 3,4′-methylenedianiline (3,4′-MDA), 3,3′-methylenedianiline (3,3′-MDA) and 3,3′-diaminobenzophenone (3,3′-DABP), using a solvent-free melt process. These imide oligomers displays low-melt viscosities (2-15 poise) at 260-280 °C, which made them amenable to low-cost resin transfer molding (RTM) process. The a-ODPA based RTM resins exhibits glass transition temperatures (Tg's) in the range of 265-330 °C after postcure at 343 °C. The mechanical properties of these polyimide/carbon fiber composites fabricated by RTM will be discussed.

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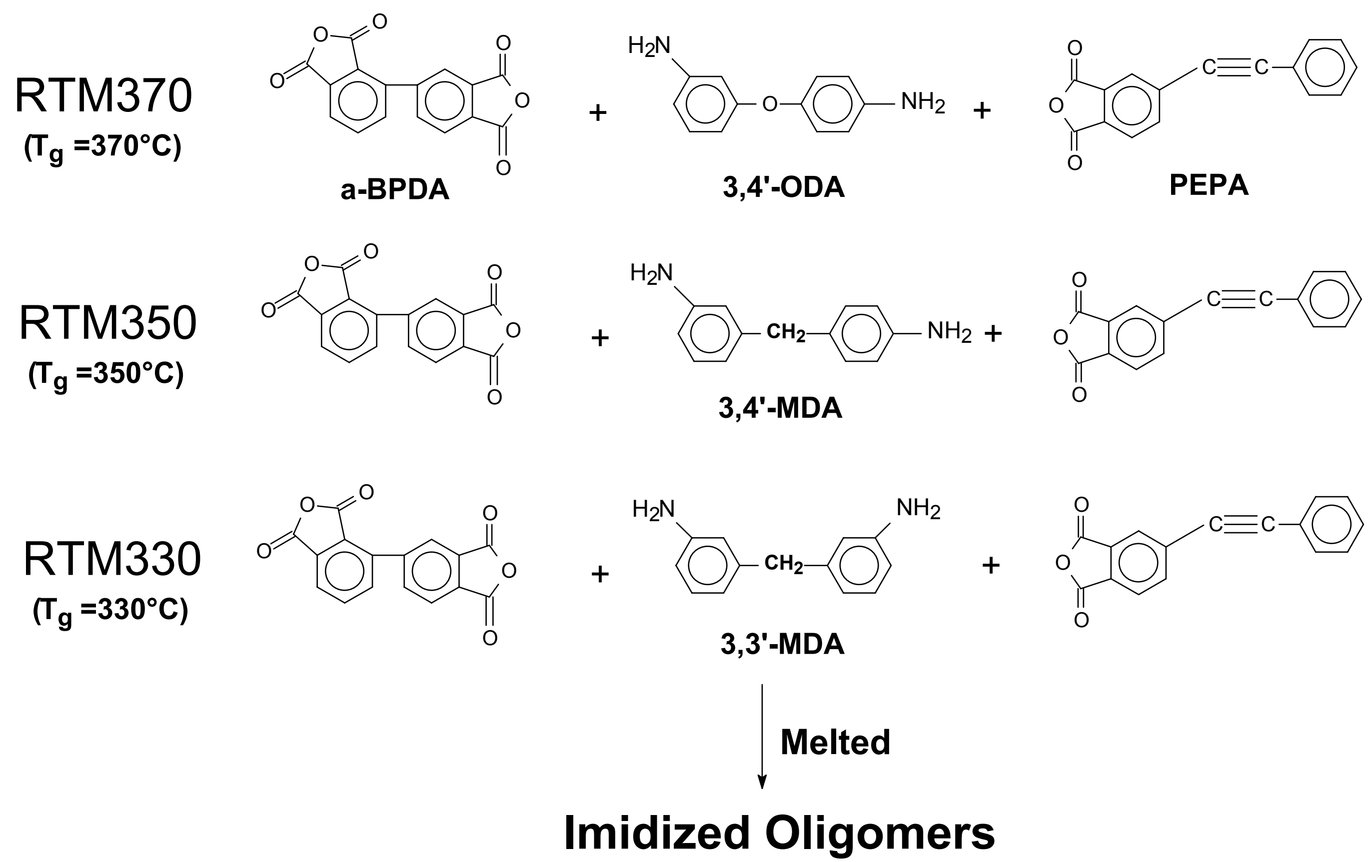
High Temperature Polyimide Composites Materials and Processing

- **PMR-15, PMR-II-50, AFR-PE4, PETI-5 composites all require solvent-based prepregs for processing**
  time consuming, costly and hazardous
- **Process polymer composites via RTM, VARTM**
  produce 30% cost saving & 12% weight saving
- **New low-melt viscosity (10-30 poise) imide resins:**
  - *amenable to low-cost RTM process*
  - *advance PMC temperature capability to 260-315°C beyond state-of the-art RTM resins, such as epoxy (177 °C) & BMI (232 °C)*

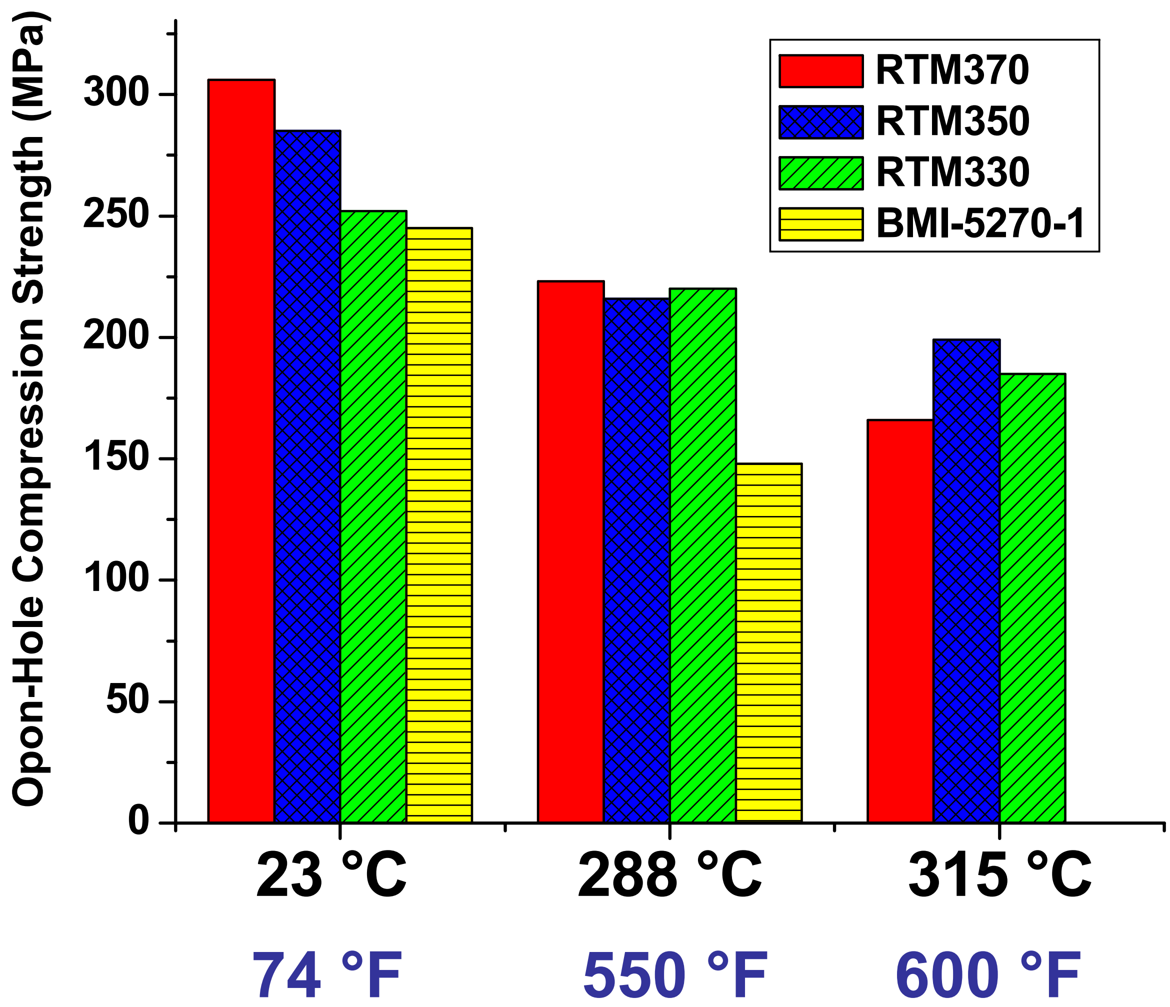
Synthesis of RTM Resins (NASA Glenn)



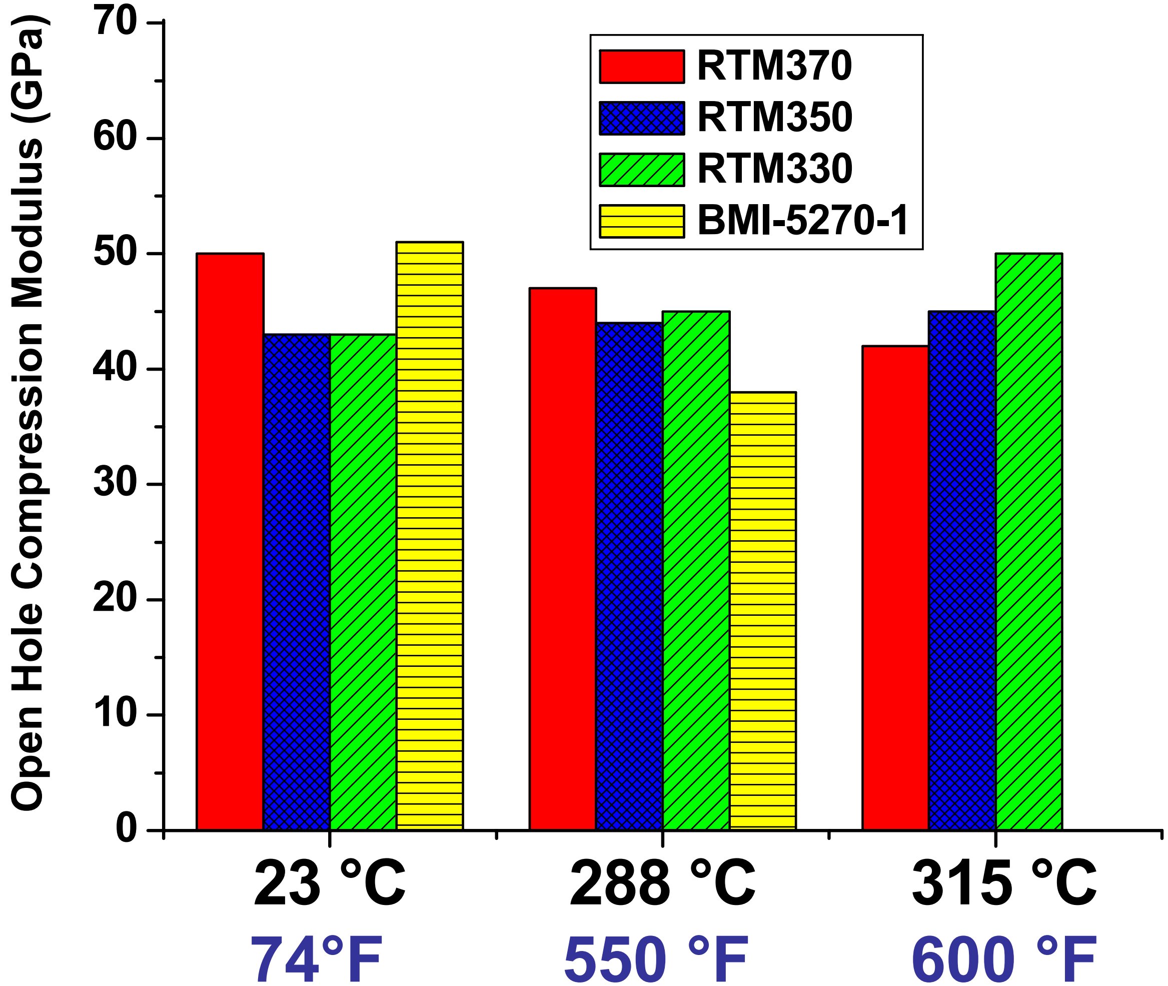
Advantages of polyimide resins containing a-BPDA vs s-BPDA

- ***Lower melt viscosities***
- ***Higher Tg's***

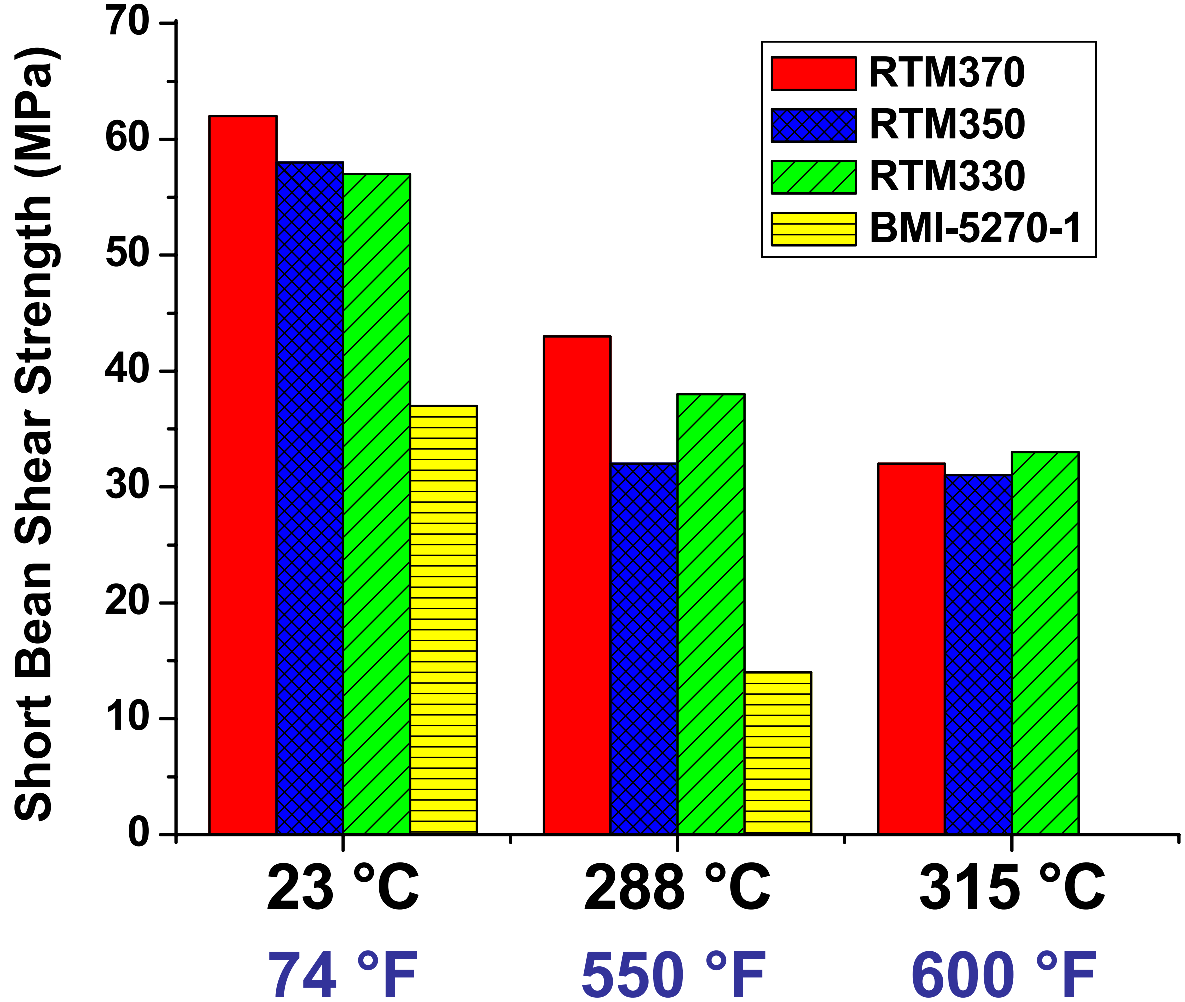
Open-Hole Compression Strength of RTM370, RTM350, RTM330 vs BMI-5270-1



Open-Hole Compression Modulus of RTM370, RTM350, RTM330 vs BMI-5270-1



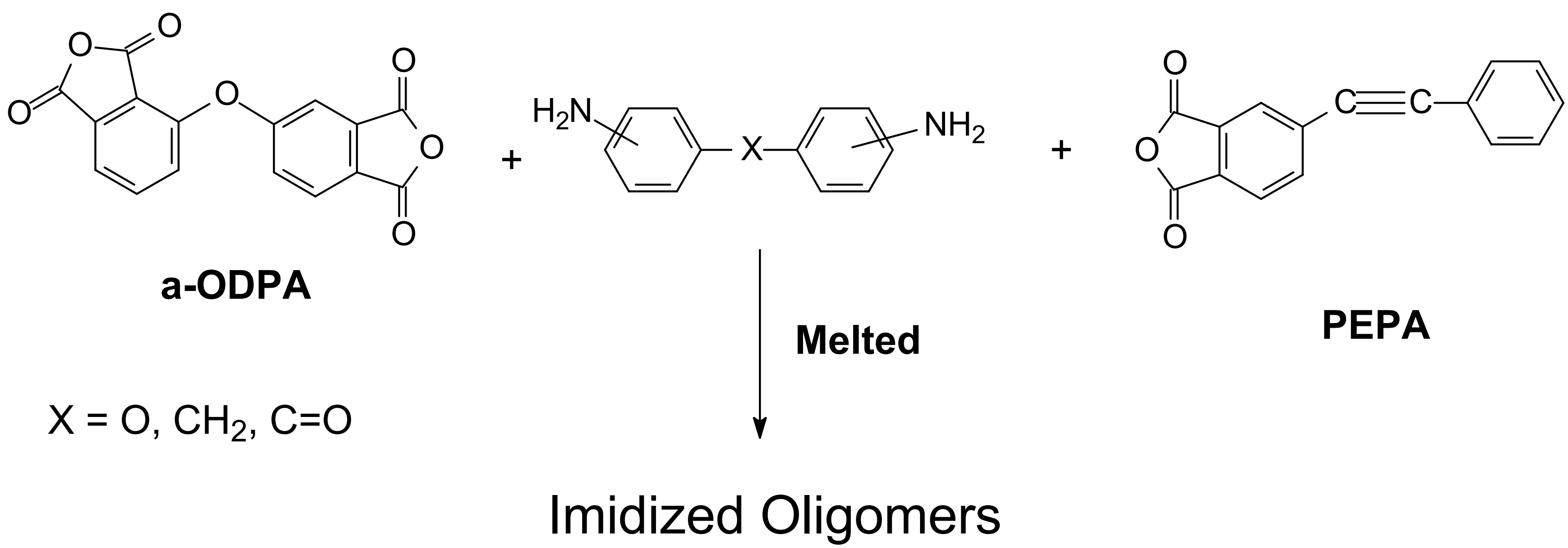
Short Beam Shear Strength of RTM370, RTM350 & RTM330 vs BMI-5270-1



New Effort in RTM Resins

- Prepare novel imide resins with low-melt viscosities (10-30 poise) that are amenable to RTM or VARTM processes
- Process new imide resins by RTM or VARTM into composite panels and evaluated mechanical properties and durability at 550-600 °F

Low-melt Viscosity Imide Resins Based on a-ODPA



Physical Properties of Imide Oligomers/Resins Based on a-ODPA / 4-PEPA

Dianhydride	Diamine	Oligomer Min. η @280 °C by Brookfield [1] (Poise)	Oligomer Min. Complex [η]* @260°C[2] (Poise)	Cured Resin T_g (°C) NPC[3] By TMA	Cured Resin T_g (°C) PC[4] @ 650°F By TMA[5]
a-ODPA	3,4′-ODA	3.5	15.0	296	329
a-ODPA	3,4′-MDA	4.0	14.0	270	294
a-ODPA	3,3′-MDA	2.5	3.0	273	266
a-ODPA	3,3′-DABP	3.0	4.0	270	297

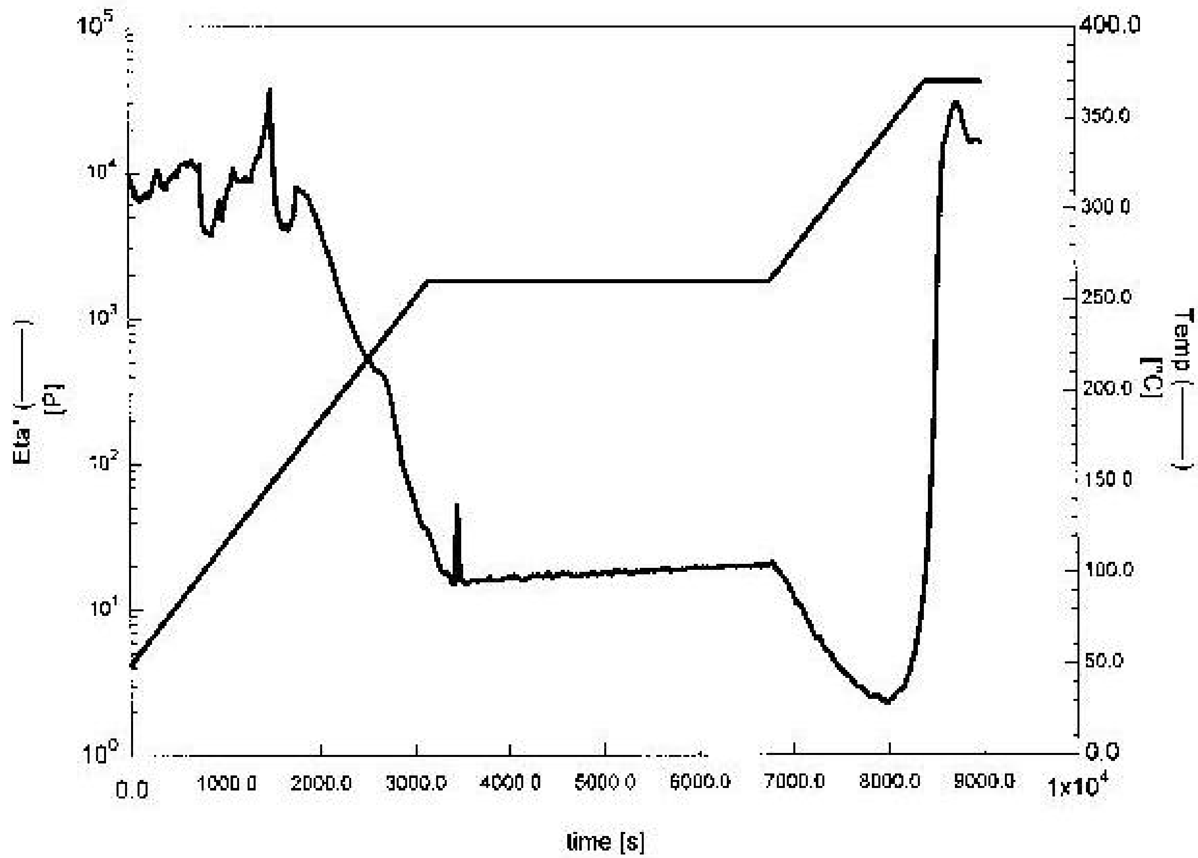
[1] Absolute viscosity measured by Brookfield Viscometer at 280 °C.
[2] Complex viscosity measured by Aries Rheometer, using parallel plates.
[3] NPC = No Post cure
[4] PC = Post cured at 343 °C (650 °F) for 16 hrs.
[5] TMA =Thermal mechanical analysis heated at 10 °C/min, using expansion mode.

Physical Properties of Imide Oligomers/Resins Based on *a-BPDA* and 4-PEPA

Resin	Diamine	Oligomer Min. η @280 °C by Brookfield [1] (Poise)	Oligomer Min. Complex [η]* @280°C [2] (Poise)	Cured Resin T_g (°C) NPC [3] byTMA	Cured Resin T_g (°C) PC[4]@ 650°F By TMA[5]
RTM370	3,4′-ODA	14	11	342	370
RTM350	3,4′-MDA	7.4	20	338	350
RTM330	3,3′-MDA	1.5	10	288	330

[1] Absolute viscosity measured by Brookfield Viscometer at 280 °C.
[2] Complex viscosity measured by Aries Rheometer, using parallel plates.
[3] NPC = No Postcure
[4] PC = Postcured at 343 °C (650 °F) for 16 hrs.
[5] TMA =Thermal mechanical analysis heated at 10 °C/min, using expansion mode.

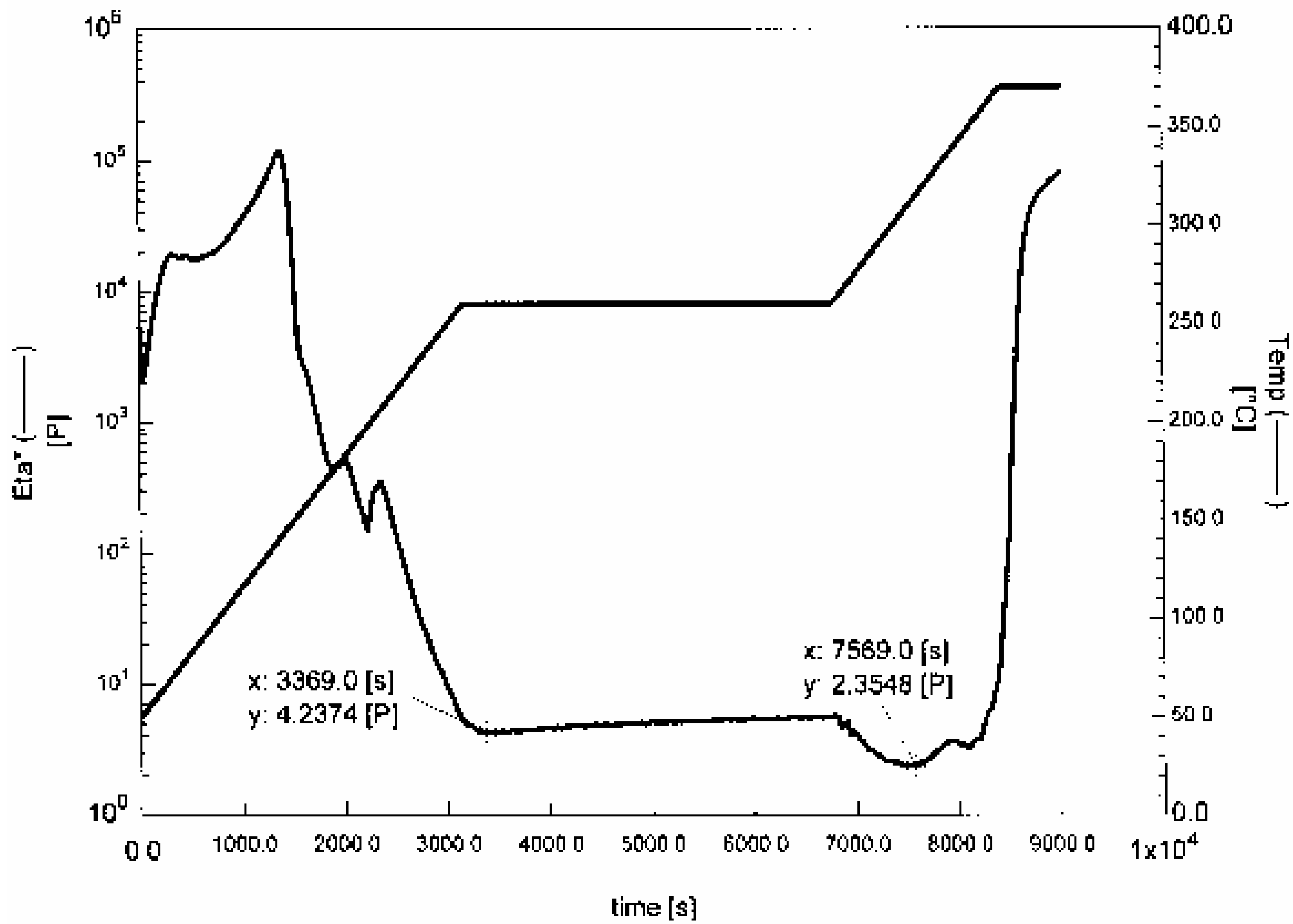
Rheology of a-ODPA/3,4′-ODA/PEPA Imide Resins at 260 °C Hold



Advantages:

Maintained low-melt viscosity (4-15 poise) at 260 °C

Rheology of a-ODPA/3,3′-DABP/PEPA Imide Resins at 260 °C Hold



Advantages:

Maintained low-melt viscosity (4-15 poise) at 260 °C

Conclusions

- **a-ODPA based RTM imide resins exhibit low melt viscosities at 260 °C comparable to a-BPDA based resins at 280 °C (10 fold)**
- **a-ODPA based RTM imide resins exhibit lower T_g's (40- 65 °C lower) than a-BPDA based RTM imide resins**

  Reason: *Additional flexible –O– linkage*

  versus

  Steric hindrance of biphenyl unit

Continued Efforts

- ♦ Fabricate composite panels from a-ODPA imide resins by RTM at 260 °C and VARTM, if feasible
- ♦ Evaluate Mechanical properties of a-ODPA/PEPA based imide composites

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