

Determining the Processability of Multilayer Coextruded Structures

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ABSTRACT

Multilayer coextrusion is a process in which two or more polymers are extruded and joined together in a feedblock or die to form a single structure with multiple layers. This paper will discuss techniques for measuring experimental rheology data for monolayer and multilayer structures and how that data can be used for determining the processability of multilayer coextruded structures.

INTRODUCTION

Many polymers are extruded through various styles of dies to produce monolayer and multilayer products. Coextrusion is a common method used for producing multilayer structures. Coextrusion is a process in which two or more polymers are extruded and joined together in a feedblock or die to form a single structure with multiple layers. This technique allows the processor to combine the desirable properties of multiple polymers into one structure with enhanced performance characteristics. The coextrusion process has been widely used to produce multilayer sheet, blown film, cast film, tubing, wire coating, and profiles (1-6).

This paper will discuss techniques for measuring experimental rheology data for monolayer and multilayer structures. Experimental data will be shown on the viscosity of single polymer and coextruded polymer melts measured using a unique rheometer for coextruded structures. These data will then be used show how the processability of coextruded structures is affected by the viscosities of the individual layers and their placement in the structure.

BACKGROUND

Many different types of monolayer and coextruded polymeric films and sheets are currently produced using different styles of dies. However, analyzing the flow of polymer melts through dies can be difficult due to the complex three dimensional flow patterns that exist (7-10). This analysis becomes even more complex when multiple layers of different materials are introduced into the structure through coextrusion methods (11-17).

One of the difficulties in designing a new coextrusion die or analyzing the flow in an existing coextrusion die is determining the rheology of the coextruded structure. No commercial equipment is available to measure the rheology of a complex coextruded structure. The purpose of this work was to experimentally measure the rheology of monolayer and coextruded structures and to show how that information can be used for determining the processability of multilayer coextruded structures.

EXPERIMENTAL

Three commercially available low density polyethylene (LDPE) resins manufactured by The Dow Chemical Company were used in the experiments: LDPE 722, LDPE 641I, and LDPE 662I. These resins will subsequently be referred to by letter designations to simplify the notation of the coextruded structures. These letter designations are Resin A for LDPE 722, Resin B for LDPE 641I, and Resin C for LDPE 662I.

Resins A, B, and C were chosen because they have significantly different viscosities based on their melt index (MI) values. Resin A has a melt index of 8, Resin B has a melt index of 2, and Resin C has a melt index of 0.47.

Two types of experiments were run in this study. The first series of experiments were run to determine the viscosity of the individual resin components. The second series of experiments were run to determine the viscosity of coextruded structures composed of combinations of the individual resins.

The coextrusion line used in this study for determining the viscosity of coextruded structures consisted of a 31.75 mm (1.25 inch) diameter, 24:1 L/D single screw extruder and a 25.4 mm (1.0 inch) diameter, 24:1 L/D single screw extruder. A schematic diagram of the extrusion line set-up is shown in Figure 1. The extruders fed individual gear pumps to ensure uniform flow of the polymer melts to the feedblock and rheology dies. The gear

pumps were attached to a feedblock by transfer lines that contained variable depth thermocouples to ensure consistent and uniform temperatures from the extruders. Several feedblocks were designed to produce coextruded structures similar to those shown schematically in Figure 2. These feedblocks allow evaluation of various coextruded structures with different layer arrangements and thicknesses.

Examples of coextruded encapsulated structures are shown in Figure 3. Black and white pigments were added to these samples to show the skin and core layer thicknesses. For the experiments in which the rheology of the structures was actually measured, no pigments were added. Figure 3 shows a sample with a relatively thin skin layer on the left and a thicker skin layer on the right.

Attached to the exit of the feedblock was one of two rheology dies that were fabricated with differing cross-sectional shapes. The first die had a circular cross section with a diameter of 7.9 mm (0.312 inch) and the second die had a rectangular cross section with a width of 25.4 mm (1.0 inch) and a height of 2.54 mm (0.1 inch). Each of these dies had four pressure transducers spaced at 50.8 mm (2.0 inch) intervals down the length of the die. Figure 4 shows photographs of the rectangular (or slit) die on the left and the circular (or rod) die on the right. The rod die is shown with the pressure transducers and the heating jacket installed.

For the rheology experiments, the coextrusion line was run with the two extruders and gear pumps producing set flow rates until steady-state conditions had been reached. At this condition, the pressures from the four transducers were recorded along with the measured total polymer flow rate and temperature. This procedure was repeated at several different flow rates so that the viscosity of the monolayer or coextruded structures could be determined at different shear rates and/or layer ratios.

RESULTS

The experimental setup shown in Figure 1 was first run with each of the three individual polyethylene resins (Resins A, B, and C). Figure 5 shows the viscosity vs. shear rate data generated for the three polyethylene resins processed at 190 C. These data show that these resins are significantly different in the magnitude of their viscosities at comparable shear rates.

The data generated for each resin were fit to a power law viscosity model and the resulting curves are shown in the Figure 5. Even though the magnitudes of the resins viscosities are significantly different at comparable shear rates, the slopes of the curves are very similar for these polyethylene resins. The slopes of these curves are related to the power law index, n . The power law indices for these resins at 190 C are 0.36, 0.35, and 0.31 for Resins A, B, and C, respectively.

For illustrative purposes, the rheology results shown and discussed in the rest of this paper will be confined to combinations of Resins A and B using the encapsulated structure shown in Figure 2.

In order to confirm that the rheology data being generated using this unique coextrusion rheometer were comparable to data generated elsewhere, these same three polyethylene resins were run offline on a capillary rheometer. Figure 6 shows a comparison of the rheology results from the coextrusion rheometer and the capillary rheometer for Resin B at 190 C. These results show that the data are consistent between the two measurement techniques and so the coextrusion rheometer should be capable of making accurate rheological measurements for comparing monolithic and coextruded structures.

The next set of experiments consisted of extruding two of the polyethylene resins (A and B) in coextruded encapsulated structures with 10% and 20% skin layers in which the skin layer was less viscous than the core layer. These experiments produced structures with 10% and 20% skin layers of Resin A on 90% and 80% cores of Resin B. The results of these experiments are shown in Figure 7 and are compared to the results for the individual resins.

These data show that the thinner the skin layer is in the structure, the more the viscosity of the structure approaches that of the core material. This result is very helpful to a die designer or troubleshooter since the coextrusion die can be designed or the flow in the die analyzed using the rheology of the skin layer to approximate the rheology of the coextruded structure if the skin layer is thick enough.

The next set of experiments consisted of extruding two of the polyethylene resins (again A and B) in coextruded encapsulated structures with 10% and 20% skin layers in which the skin layer was more viscous than the core layer. These experiments produced structures with 10% and 20% skin layers of Resin B on 90% and 80% cores

of Resin A. The results of these experiments are shown in Figure 8 and again are compared to the results for the individual resins.

These data also show that the thinner the skin layer is in the structure, the more the viscosity of the structure approaches that of the core material, similar to what was shown in Figure 7 with the skin and core materials inverted in the structure.

The final set of experiments consisted of measuring the viscosity of encapsulated structures of Resins A and B in which the skin layer thicknesses are varied from 0 to 50% of the total structure. These experiments were run at a constant shear rate of approximately 40 1/s. The results of these experiments are shown in Figures 9 and 10.

Figure 9 shows the viscosity of encapsulated structures with a skin layer of Resin A on a core of Resin B as the skin layer thickness is varied from 0 to 50% of the structure. Since Resin A is less viscous than Resin B, a decrease in the structure viscosity would be expected as a layer of Resin A is added to the structure. Figure 9 shows that there is a significant drop in the viscosity of the structure even with just a 5% skin layer of Resin A added to the structure.

Figure 10 shows the viscosity of encapsulated structures with a skin layer of Resin B on a core of Resin A as the skin layer thickness is varied from 0 to 50% of the structure. Since Resin B is more viscous than Resin A, an increase in the structure viscosity would be expected as a layer of Resin B is added to the structure. Figure 10 shows that there is a substantial increase in the viscosity of the structure as a layer of Resin B is added to the structure.

The viscosity data presented for the coextruded structures is one way to look at the processability of those structures. Another way to look at the processability of a coextruded structure is to look at the pressure drop produced by the structure as it flows through the processing equipment. For the circular (rod) die used in these experiments, the pressure drop created as the resins were extruded through the die was recorded at each flow rate and layer ratio. For example, the pressure drop for Resin A flowing through the circular die at a shear rate of 40 1/s was 13.1 MPa/m (48.2 psi/in). The pressure drop for Resin B flowing through the same die at the same shear rate was 26.9 MPa/m (99.1 psi/in).

When creating a coextruded structure, it is not only important to consider the resins used in the structure from a final physical property standpoint, it is also very important to consider the effect of the viscosity of the resins in the layers and their placement in the structure on the processability of the structure. For example, it might improve the final physical properties of a coextruded structure if a layer of Resin B was placed on the surface of a structure using Resin A as the core layer. This might make the structure more difficult to process from a pressure drop standpoint, however, since Resin B is more viscous than Resin A. Adding only a 10% layer of Resin B would increase the system pressure drop from 13.1 MPa/m (48.2 psi/in) to 18.1 MPa/m (66.5 psi/in), a 38% increase, in the equipment used in these experiments.

CONCLUSIONS

A unique apparatus has been developed to measure the rheology of coextruded structures. This apparatus has been used to measure the rheology of monolithic and coextruded encapsulated structures. The results of these experiments show that the rheology of coextruded structures is affected by the relative viscosities of the individual components, the thickness of the layers in the structure, as well as the placement of the layers in relation to each other. These findings are significant since they give a die designer or troubleshooter more information on how to design a coextrusion die or analyze the flow in the die using the rheology of the different layers to approximate the rheology of the coextruded structure.

When creating a coextruded structure, it is not only important to consider the resins used in the structure from a final physical property standpoint, it is also very important to consider the effect of the viscosity of the resins in the layers and their placement in the structure on the processability of the structure.

ACKNOWLEDGEMENTS

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Keywords: Rheology, coextrusion, encapsulation, die design.

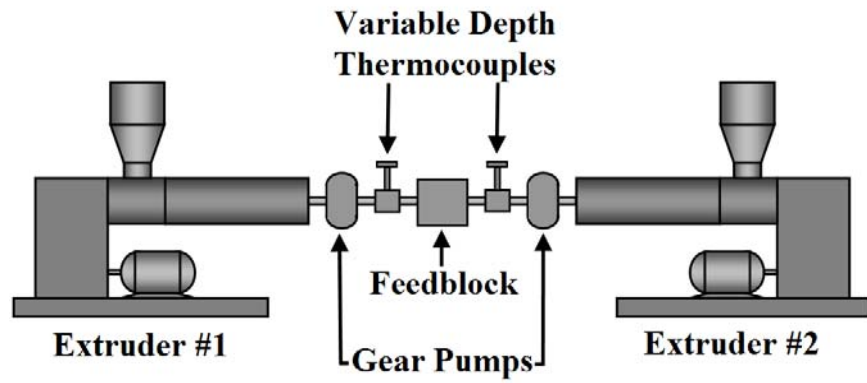


Figure 1. Coextrusion equipment setup.

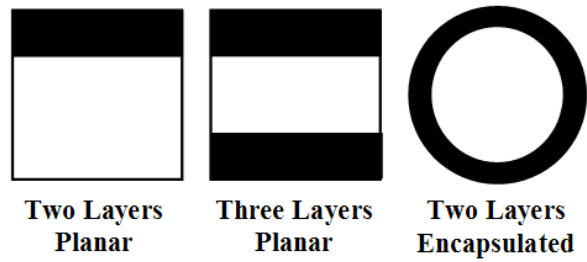


Figure 2. Coextruded structure capabilities.

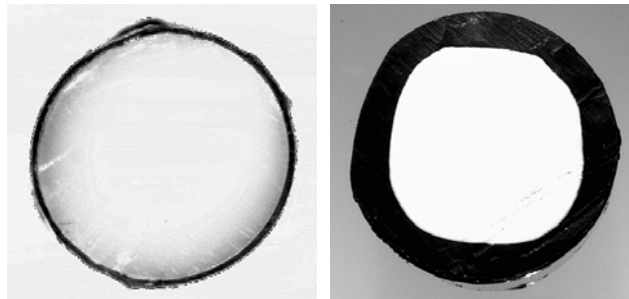


Figure 3. Coextruded encapsulated structures with different skin layer thicknesses.

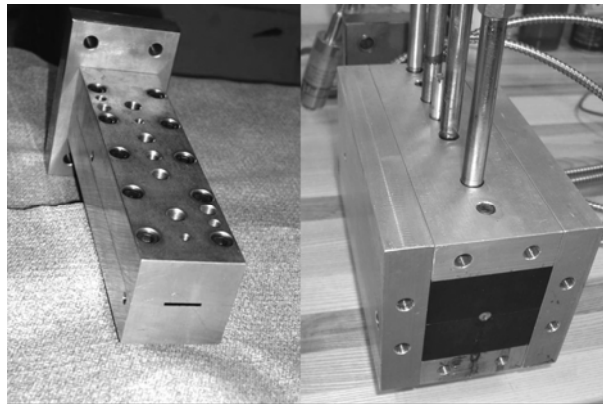


Figure 4. Dies for measuring rheology of coextruded structures.

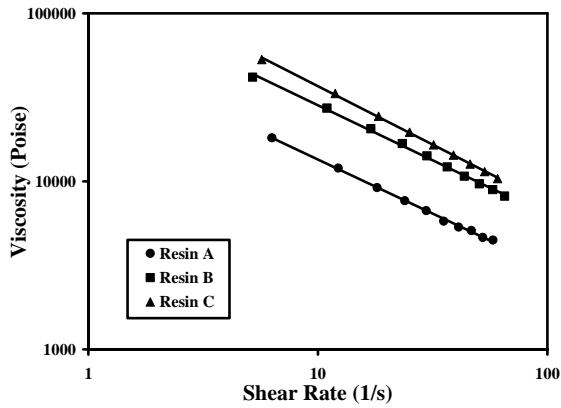


Figure 5. Comparison of the viscosity data for the three LDPE resins at 190 C.

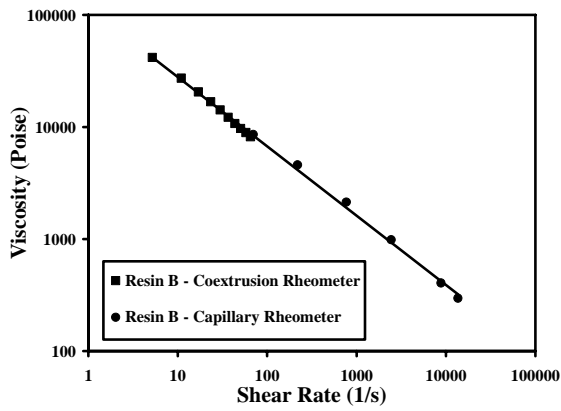


Figure 6. Comparison of the viscosity data from the coextrusion rheometer and a capillary rheometer.

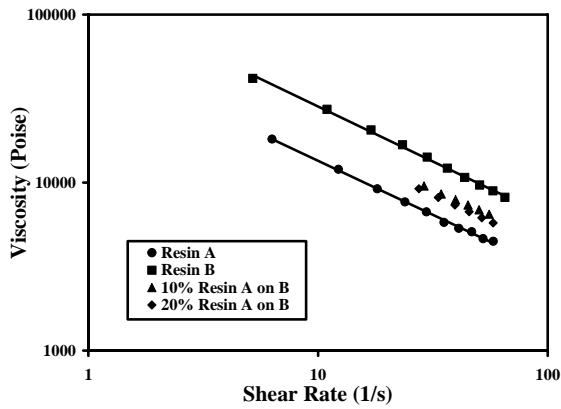


Figure 7. Viscosity data for individual components and for coextruded encapsulated structures with the skin layer being less viscous than the core layer.

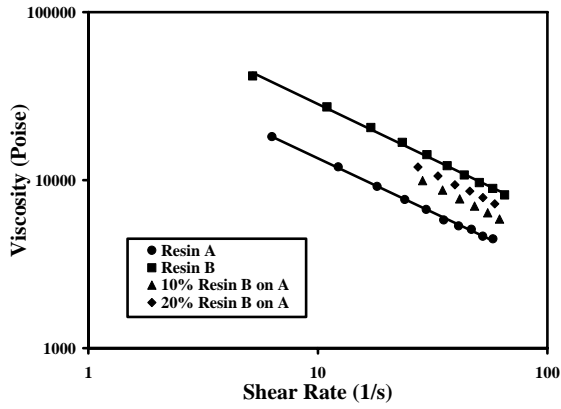


Figure 8. Viscosity data for individual components and for coextruded encapsulated structures with the skin layer being more viscous than the core layer.

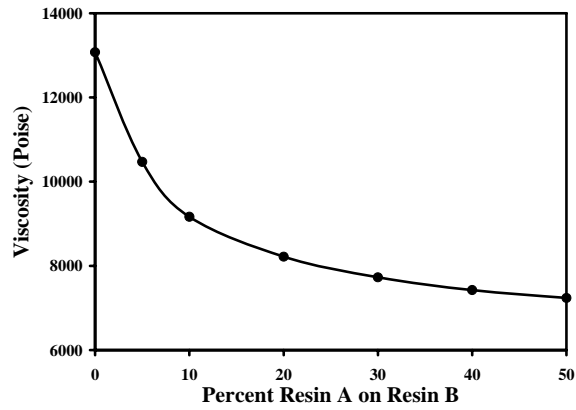


Figure 9. Viscosity data for different layer thickness percentages of Resin A on Resin B.

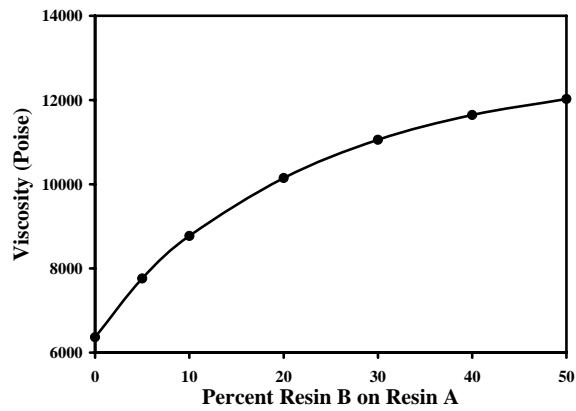


Figure 10. Viscosity data for different layer thickness percentages of Resin B on Resin A.



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Coextrusion

Coextrusion is the process of joining two or more polymer layers together in a structure to produce a product with enhanced properties

Opportunity

- **Coextruded structure designs are typically based on a desired physical property in the final product (barrier, esthetics, adhesion, etc.)**
- **The processability of these coextruded structures should also be considered since that affects the final properties and economics of the system**

Factors Affecting Processability of Coextruded Structures

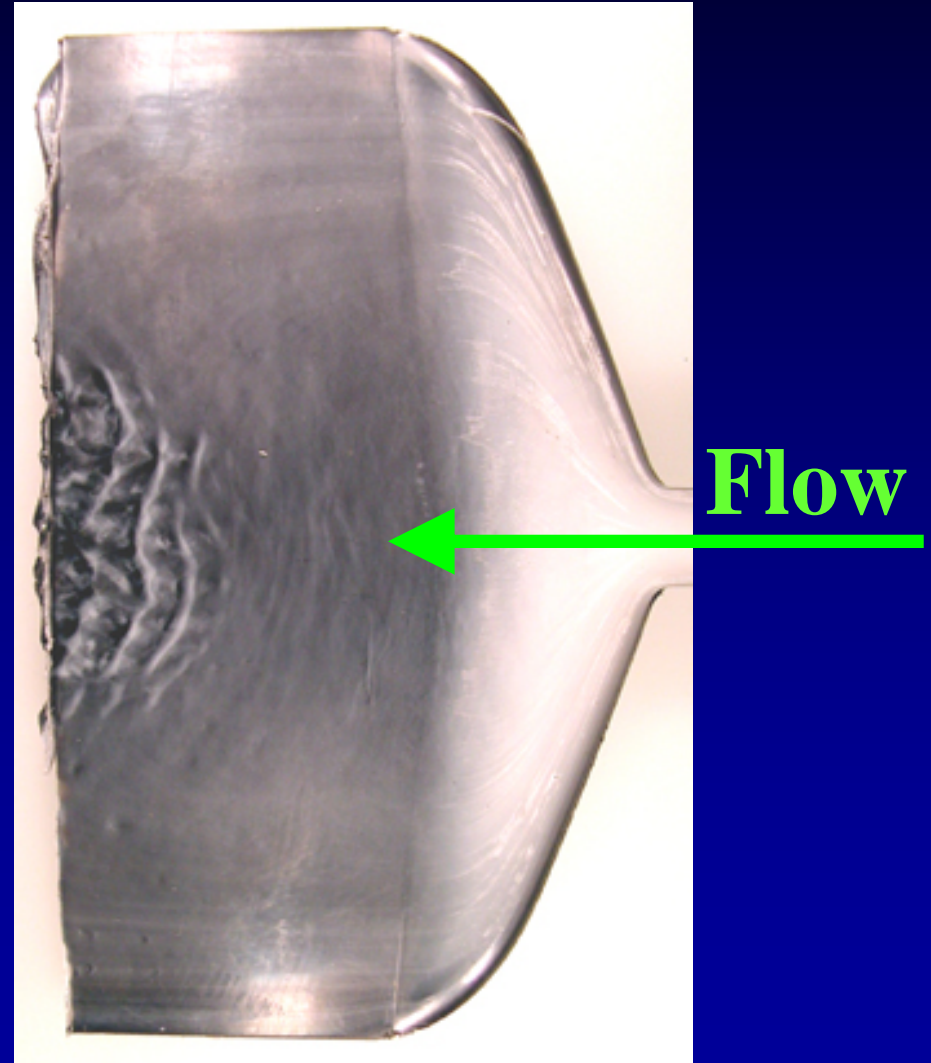
- **Processing Limitations Due to:**
 - **Interlayer Instability**
 - **Layer Uniformity**
 - **Pressure Drop**

Factors Affecting Processability of Coextruded Structures

- Processing Limitations Due to:
 - **Interlayer Instability**
 - Layer Uniformity
 - Pressure Drop

Interlayer Instability

Interlayer instability is a non-uniform, “wavy” interface between layers that can get more pronounced at higher flow rates



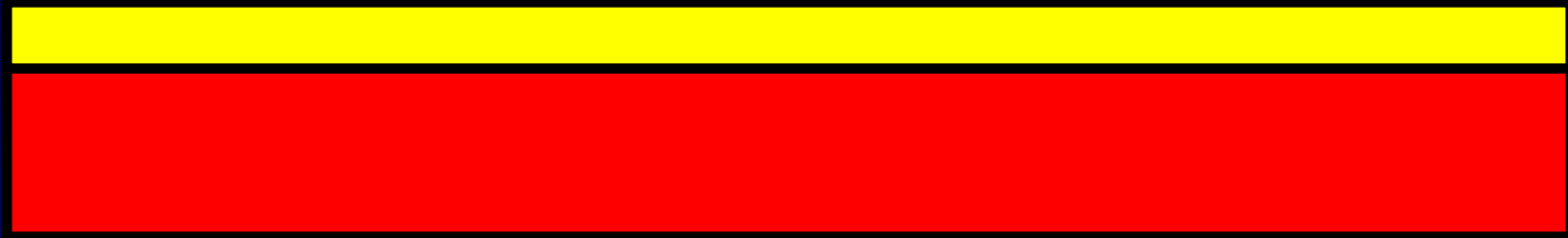
Factors Affecting Processability of Coextruded Structures

- **Processing Limitations Due to:**
 - Interlayer Instability
 - **Layer Uniformity**
 - Pressure Drop

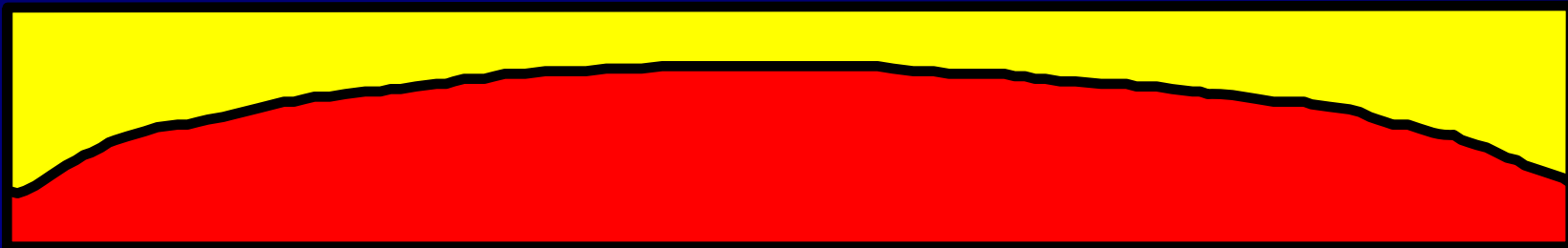
Factors Affecting Processability of Coextruded Structures

- **Processing Limitations Due to:**
 - **Interlayer Instability**
 - **Layer Uniformity**
 - **Viscous Encapsulation**
 - **Elastic Layer Rearrangement**
 - **Pressure Drop**

Layer Thickness Uniformity



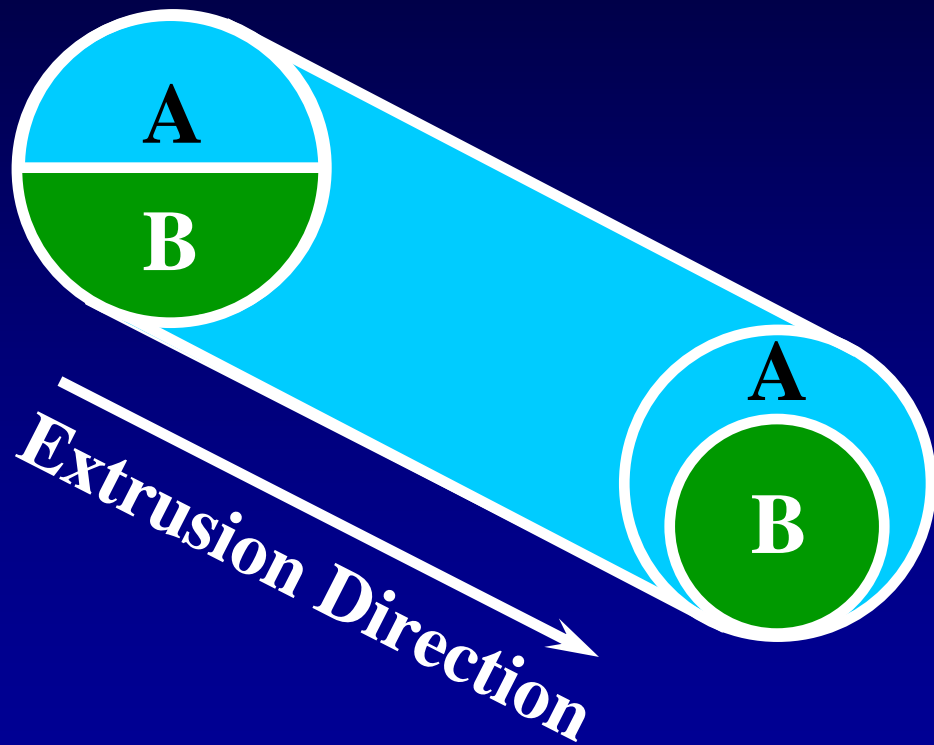
Good Layer Uniformity



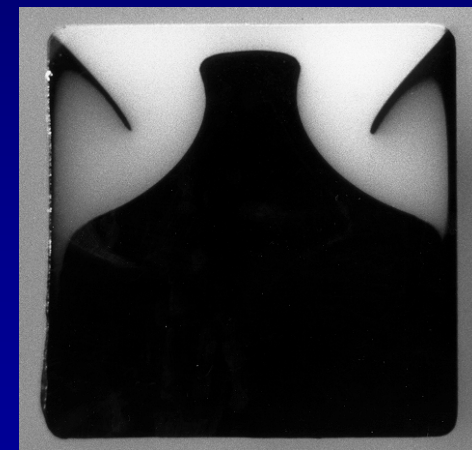
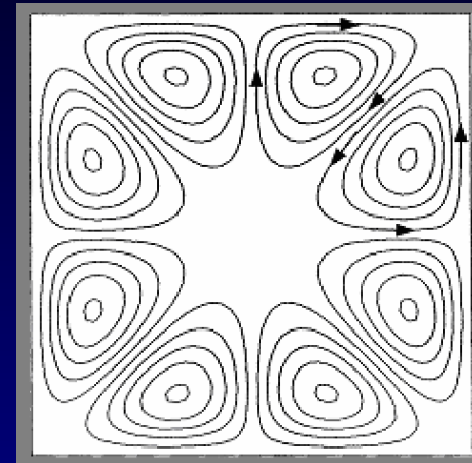
Poor Layer Uniformity

Can be caused by viscous and/or elastic effects

Layer Thickness Uniformity



Viscous Encapsulation

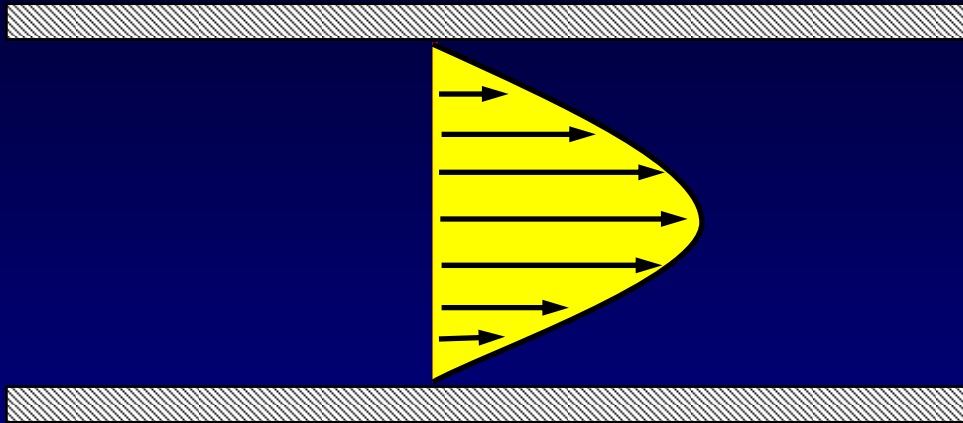


Elastic Layer Rearrangement

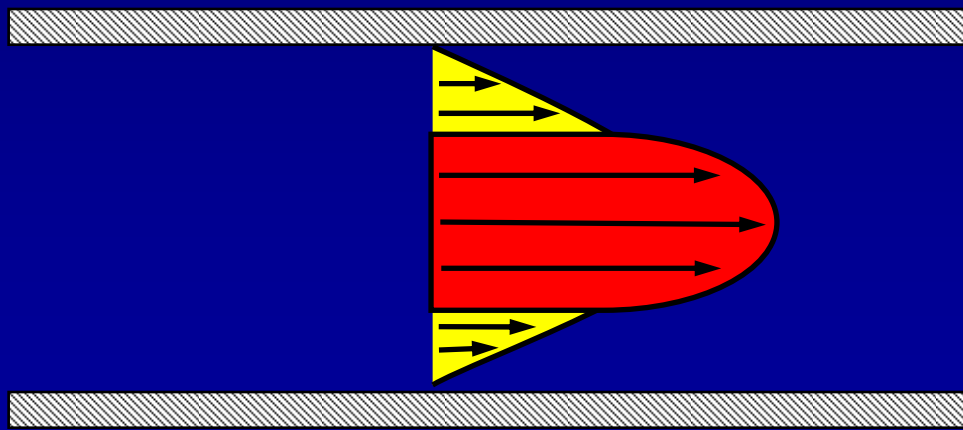
Factors Affecting Processability of Coextruded Structures

- **Processing Limitations Due to:**
 - **Interlayer Instability**
 - **Layer Uniformity**
 - **Pressure Drop**
 - **Viscosity of Coextruded Structure**
 - **Layer Viscosities**
 - **Layer Placement**
 - **Layer Thickness**

Pressure Drop



Single Material



Coextruded
Material

Pressure Drop Equations

Tube:
$$\Delta P = \frac{8Q\eta L}{\pi R^4}$$

Slit:
$$\Delta P = \frac{12Q\eta L}{wh^3}$$

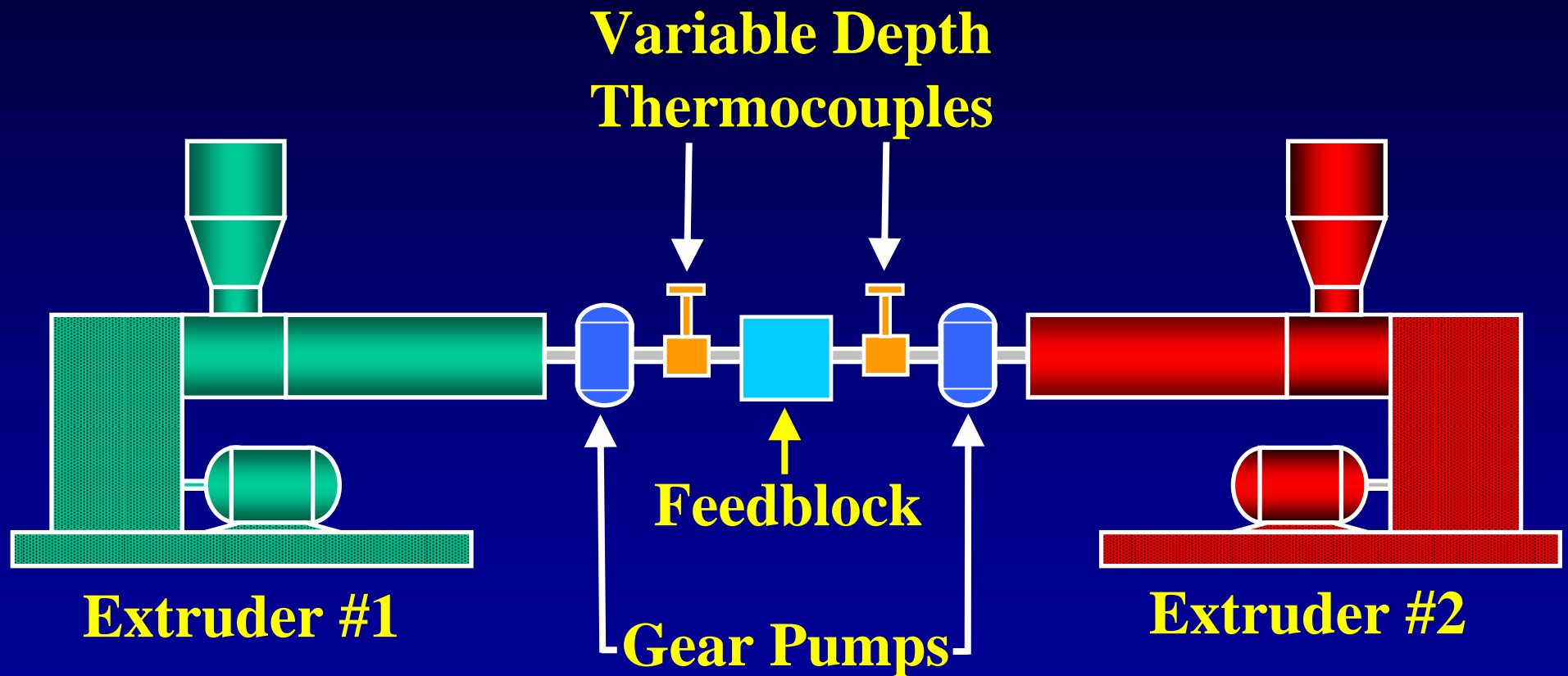
Pressure Drop Equations

Tube: $\Delta P = \frac{8Q\eta L}{\pi R^4}$

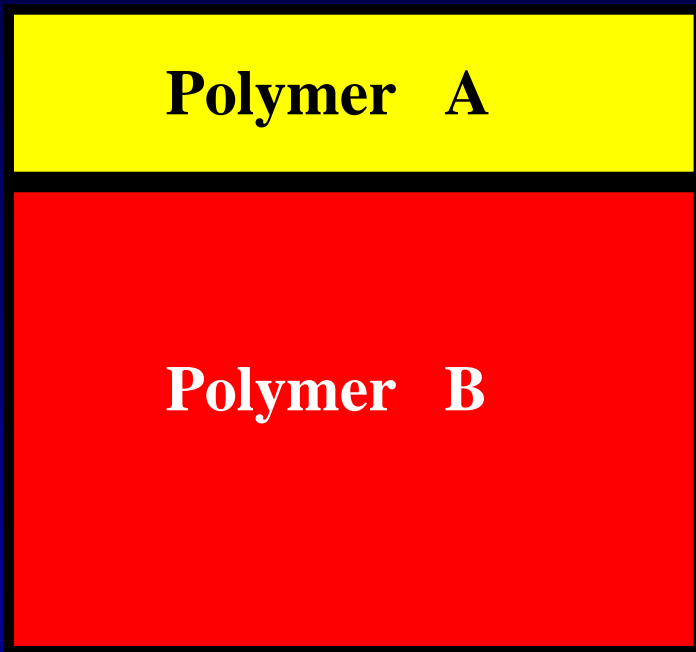
Slit: $\Delta P = \frac{12Q\eta L}{wh^3}$

**Multilayer
Rheology
Measurement**

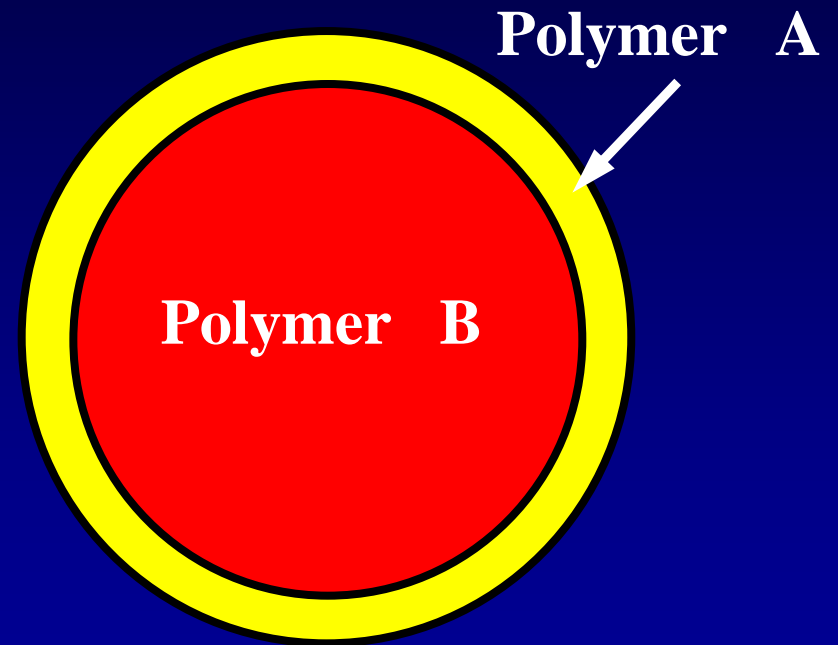
Coextrusion Rheology Equipment



Feedblock Types

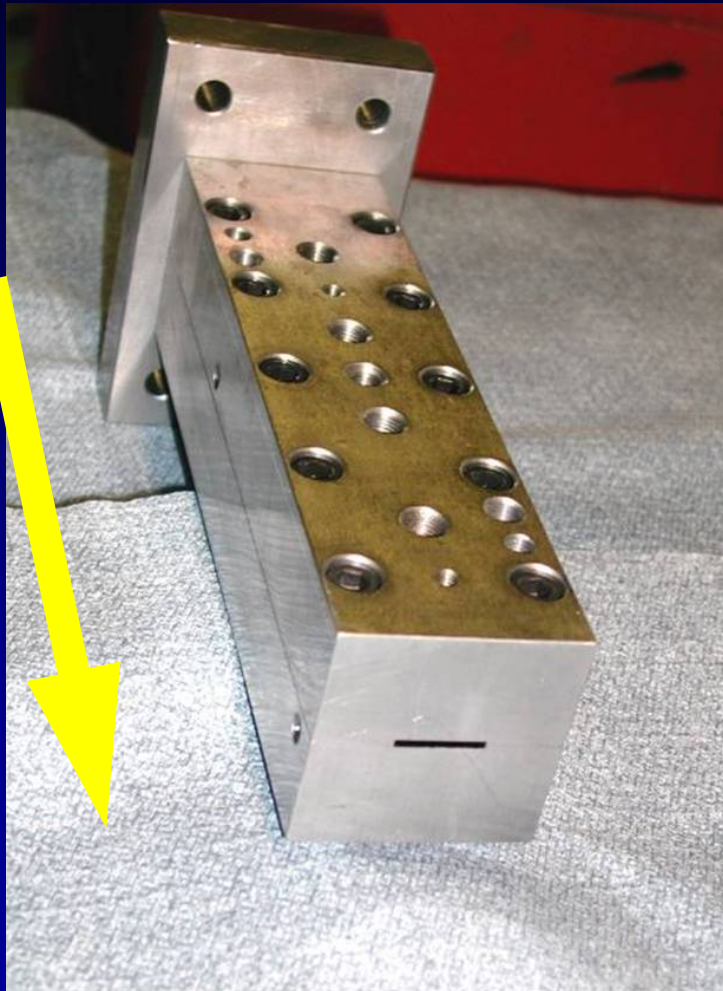


Planar Layers



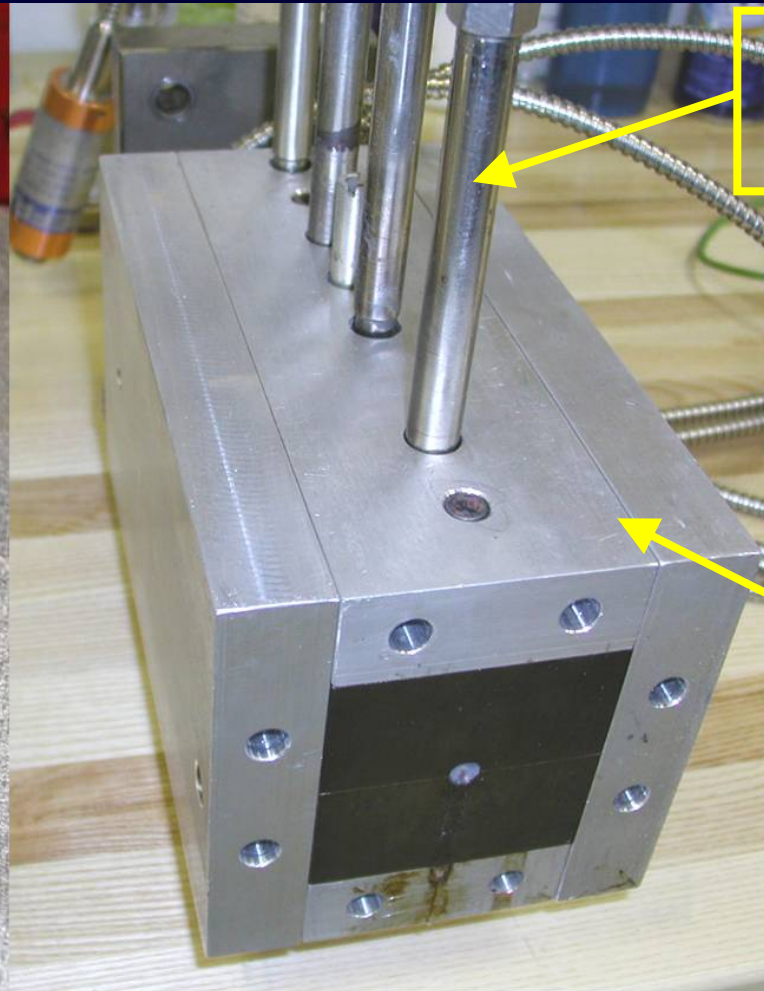
Encapsulated Layers

Rheology Die Types



**Flow
Direction**

Slit Die

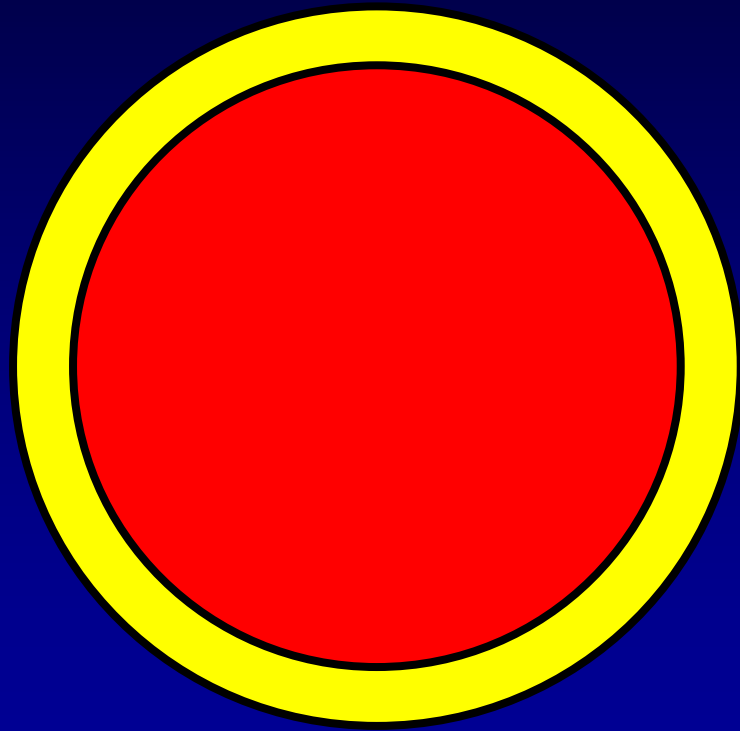


**Pressure
Transducer**

**Heating
Jacket**

Rod Die

Feedblock/Die Combination



Encapsulated Structure



Rod Die

Materials

Materials

<u>Resin Type</u>	<u>Designation</u>	<u>MI</u>
Polyethylene	Resin A	8
Polyethylene	Resin B	2
Polyethylene	Resin C	1
Polyethylene	Resin D	5

Power Law Viscosity Model

$$\eta = K \dot{\gamma}^{(n-1)}$$

Viscosity **Consistency Index** **Shear Rate** **Power Law Index**

The diagram shows the equation $\eta = K \dot{\gamma}^{(n-1)}$ in white text on a dark blue background. Below the equation, four labels in yellow text are connected to their respective parts of the equation by yellow arrows. The label 'Viscosity' has an arrow pointing to the Greek letter η . The label 'Consistency Index' has an arrow pointing to the letter K . The label 'Shear Rate' has an arrow pointing to the dot over the Greek letter $\dot{\gamma}$. The label 'Power Law Index' has an arrow pointing to the exponent $(n-1)$.

Materials

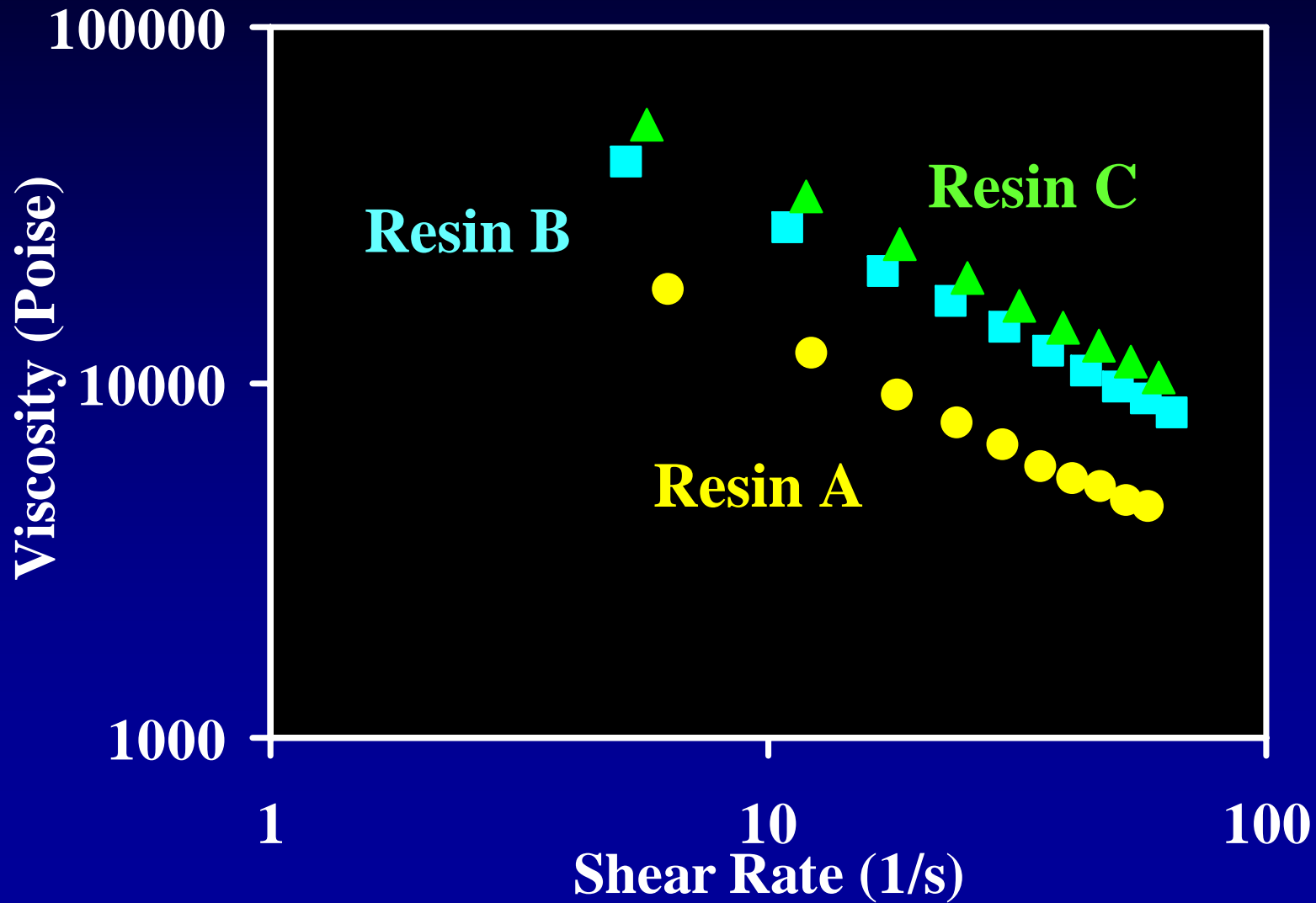
<u>Resin Type</u>	<u>Designation</u>	<u>MI</u>	<u>“n”</u>
Polyethylene	Resin A	8	0.36
Polyethylene	Resin B	2	0.35
Polyethylene	Resin C	1	0.31
Polyethylene	Resin D	5	0.55

Rheometer Evaluation

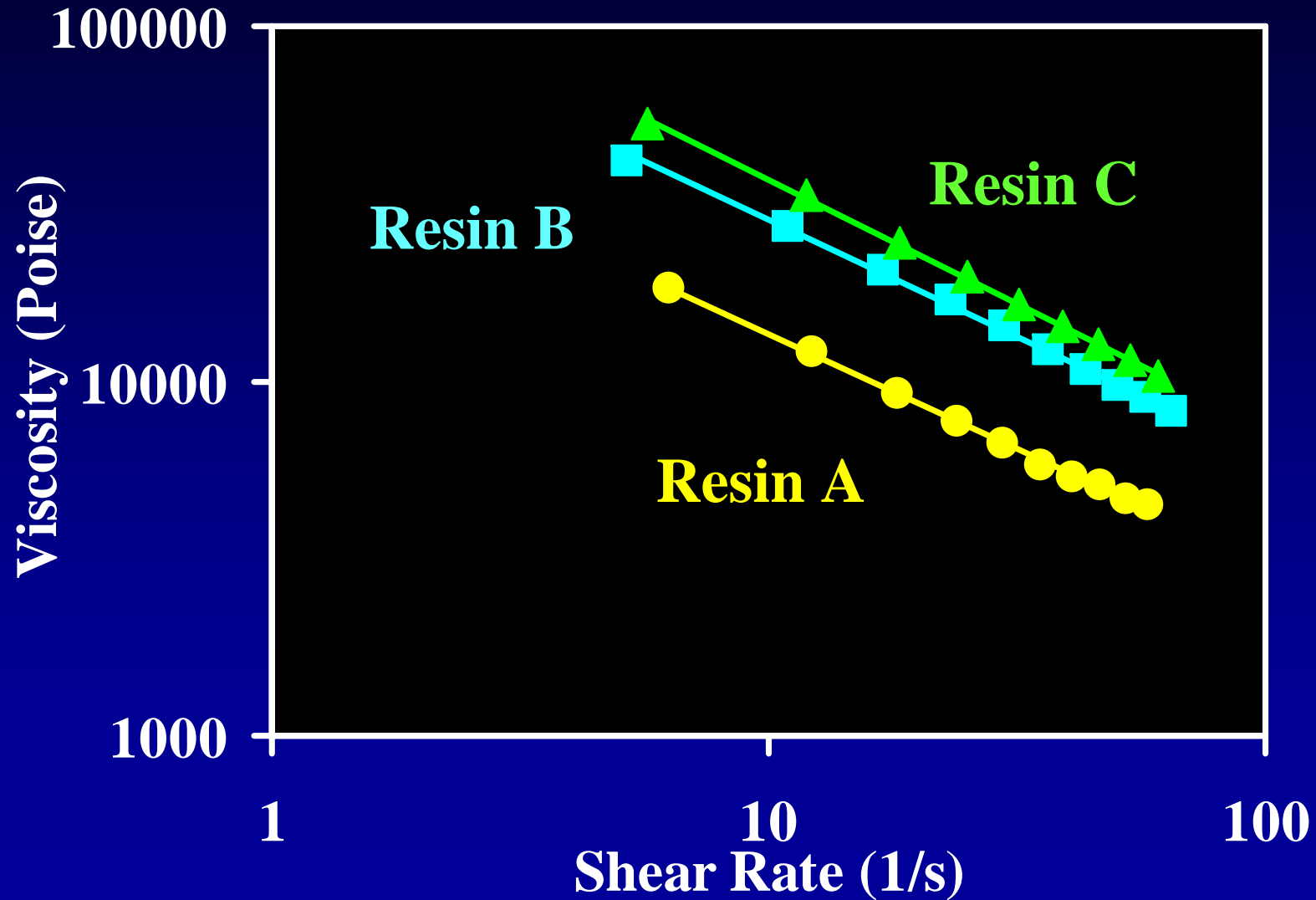
Materials

<u>Resin Type</u>	<u>Designation</u>	<u>MI</u>	<u>“n”</u>
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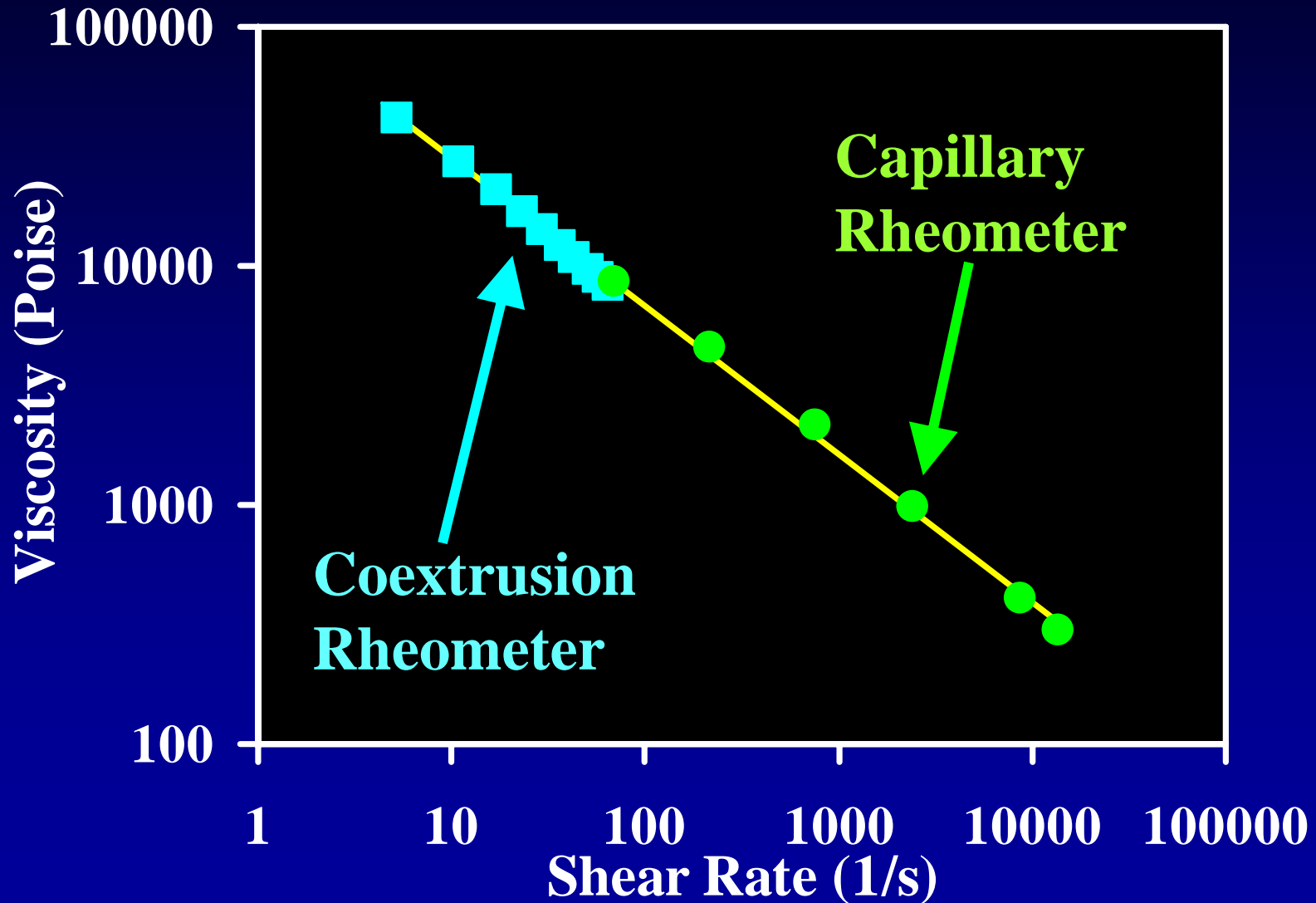
Rheometer Evaluation



Rheometer Evaluation



Rheometer Comparison for Resin B



Experimental Design

Experimental Design

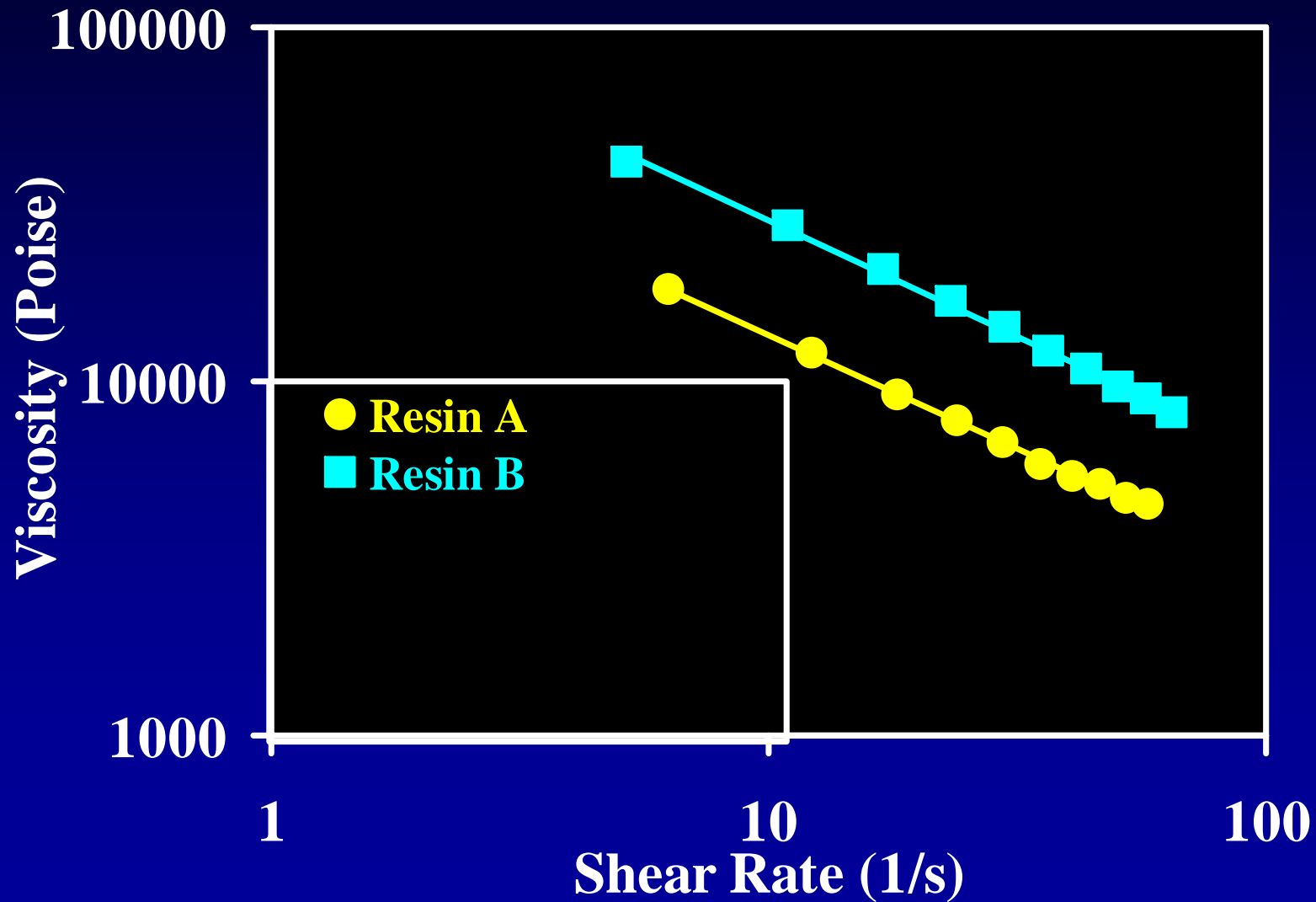
- **Layer Viscosity/Placement Effects**
 - Less viscous encapsulating more viscous
 - More viscous encapsulating less viscous
 - Resins A and B
- **Layer Thickness Effects**
 - Resins A and B
- **Shear Thinning Effects**
 - Resins A and D

Layer Viscosity Effects

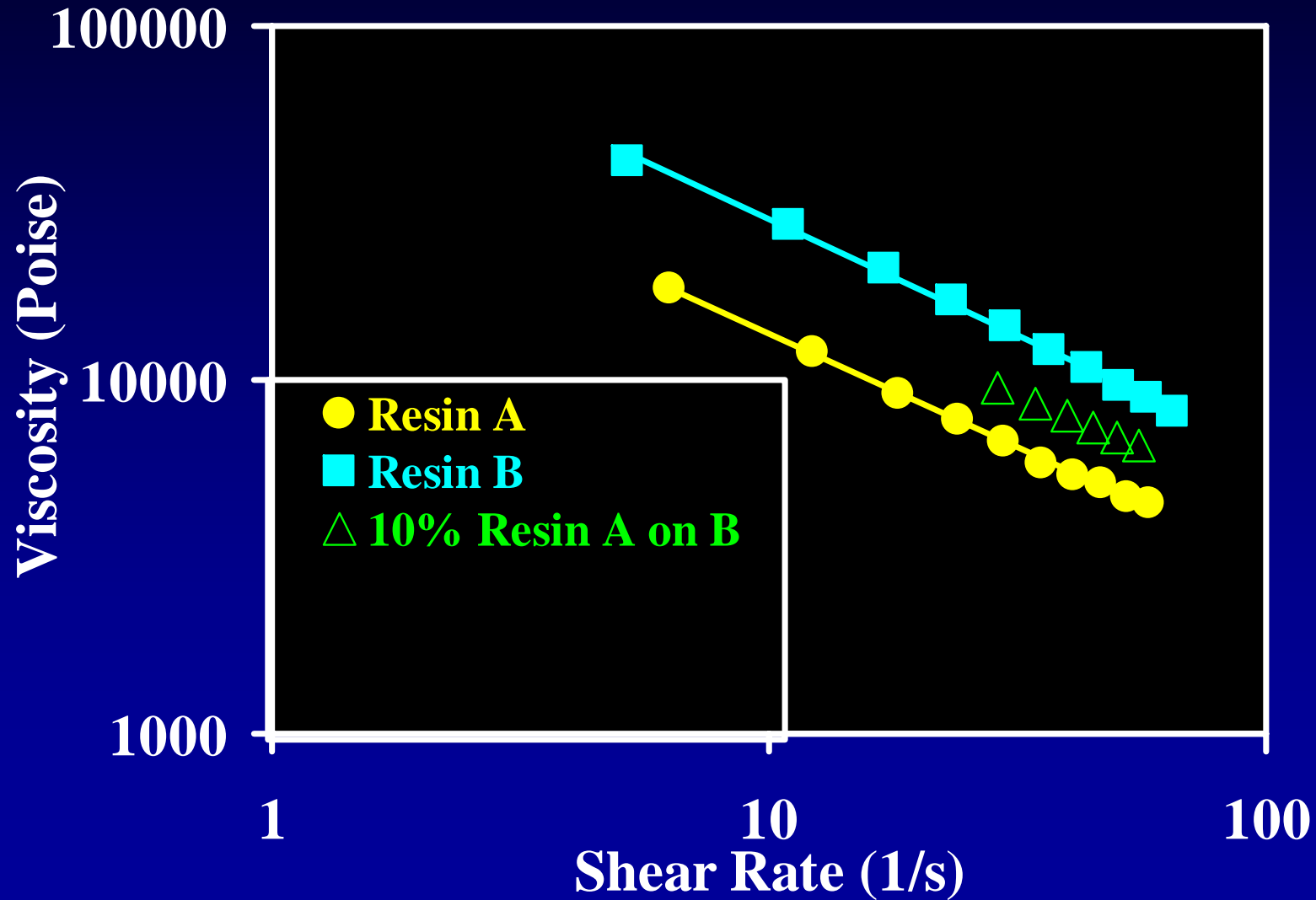
Layer Viscosity Effects

- **Measure viscosity of a coextruded structure as a function of layer viscosity and placement**
- **Produce structures with 0, 10, 20, 30, 40 and 100% skin layers**
- **Less viscous material in skin layer:**
 - **Resin A encapsulating Resin B**
- **More viscous material in skin layer:**
 - **Resin B encapsulating Resin A**

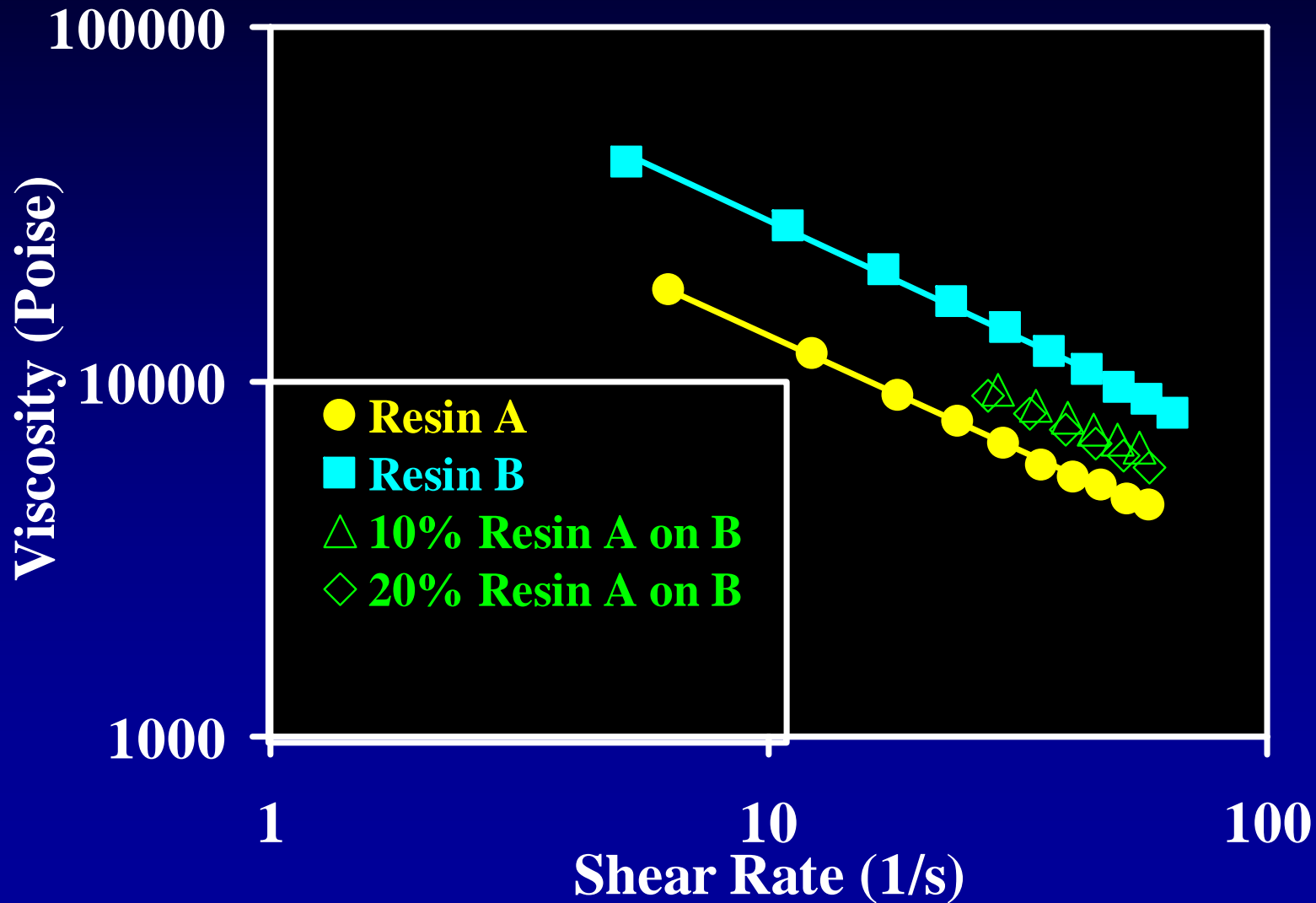
Monolayer Rheology



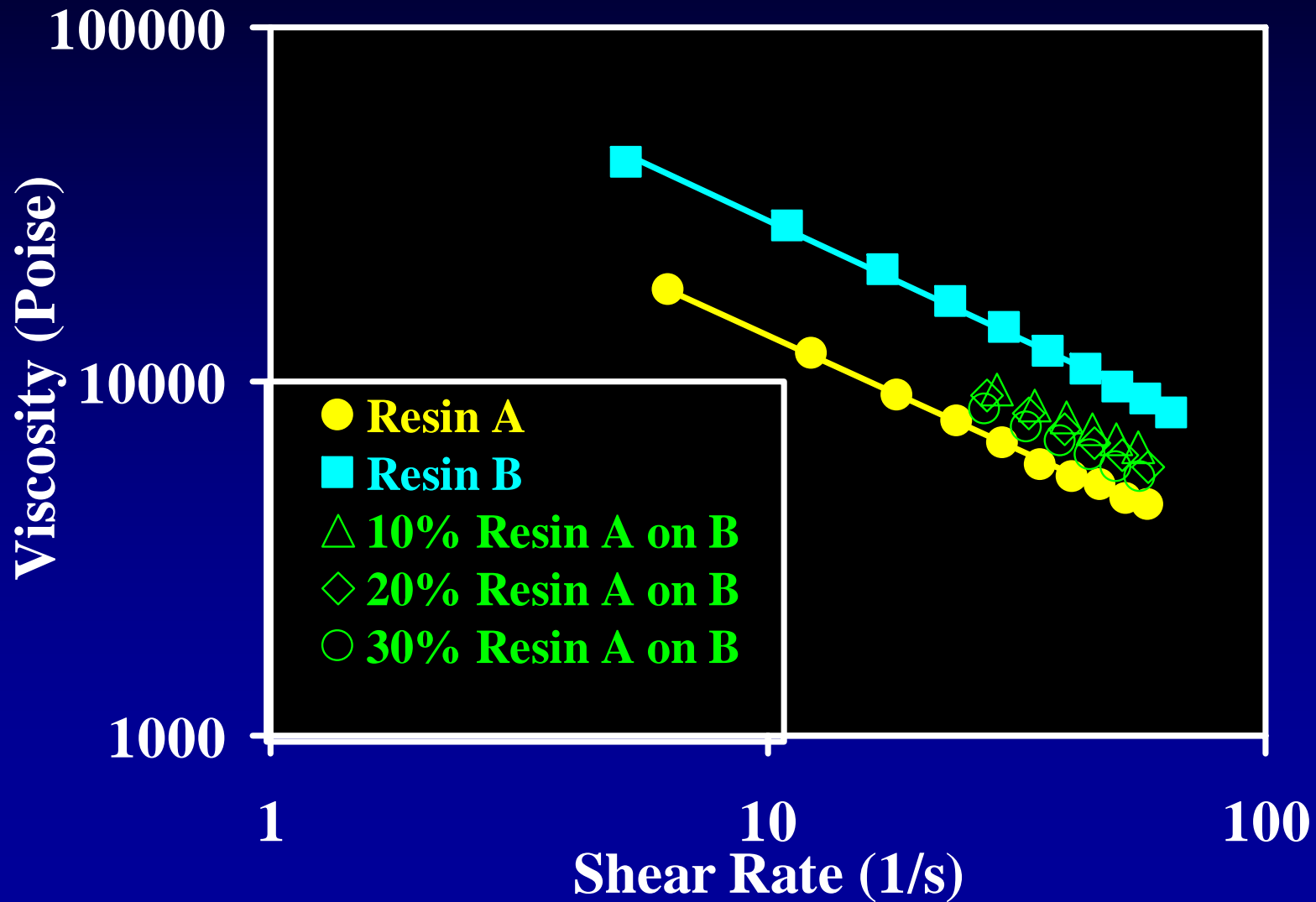
Multilayer Rheology



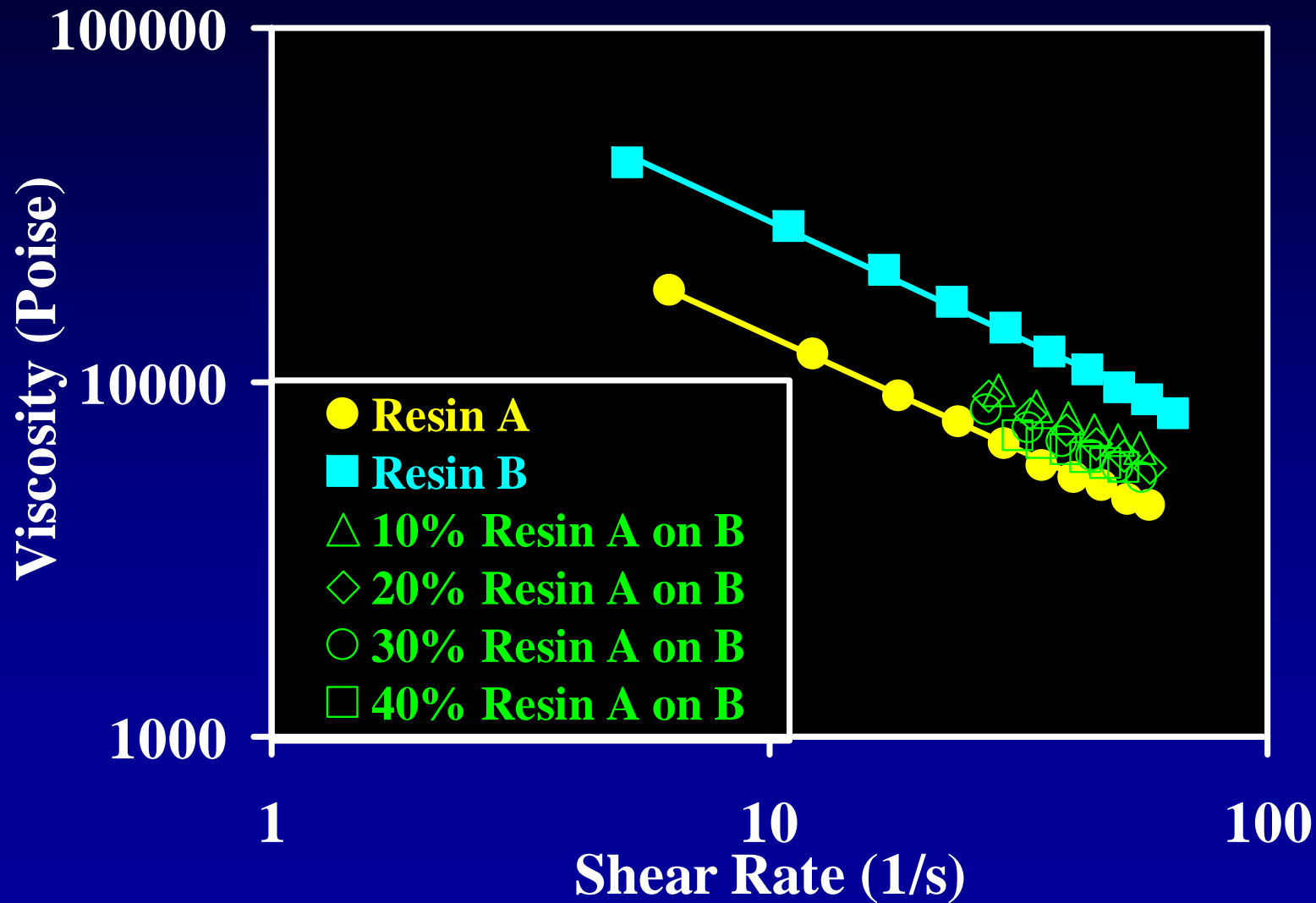
Multilayer Rheology



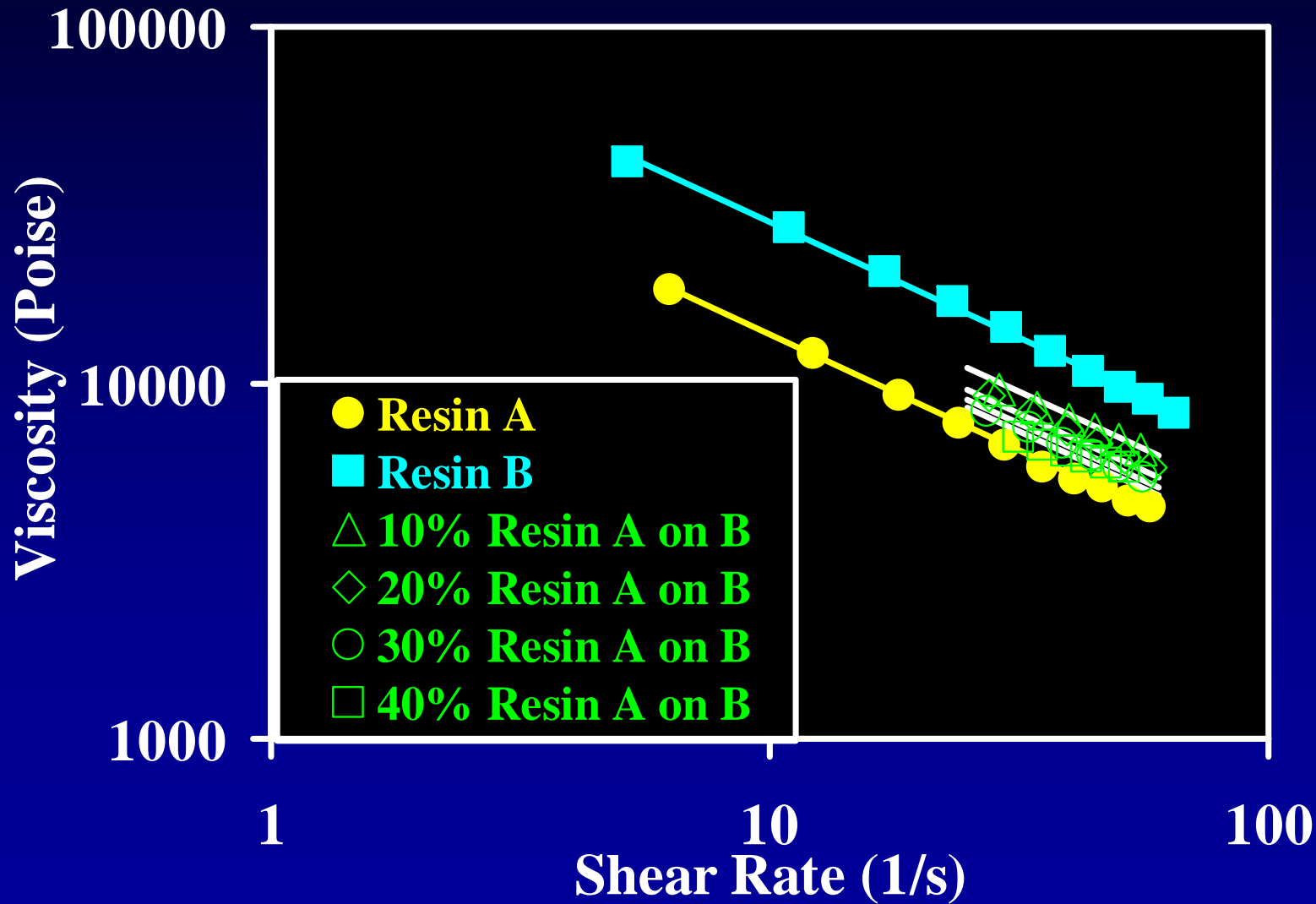
Multilayer Rheology



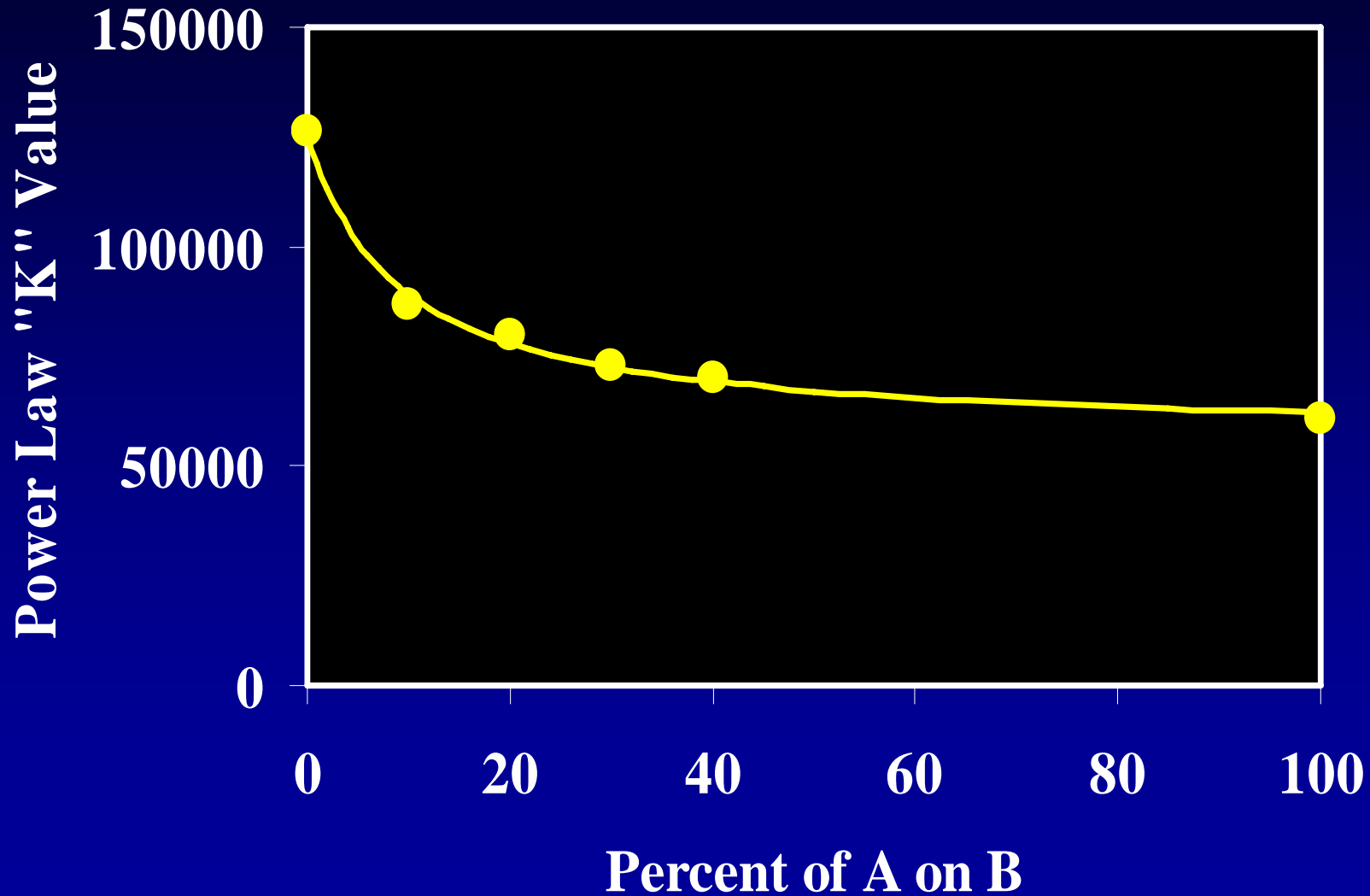
Multilayer Rheology



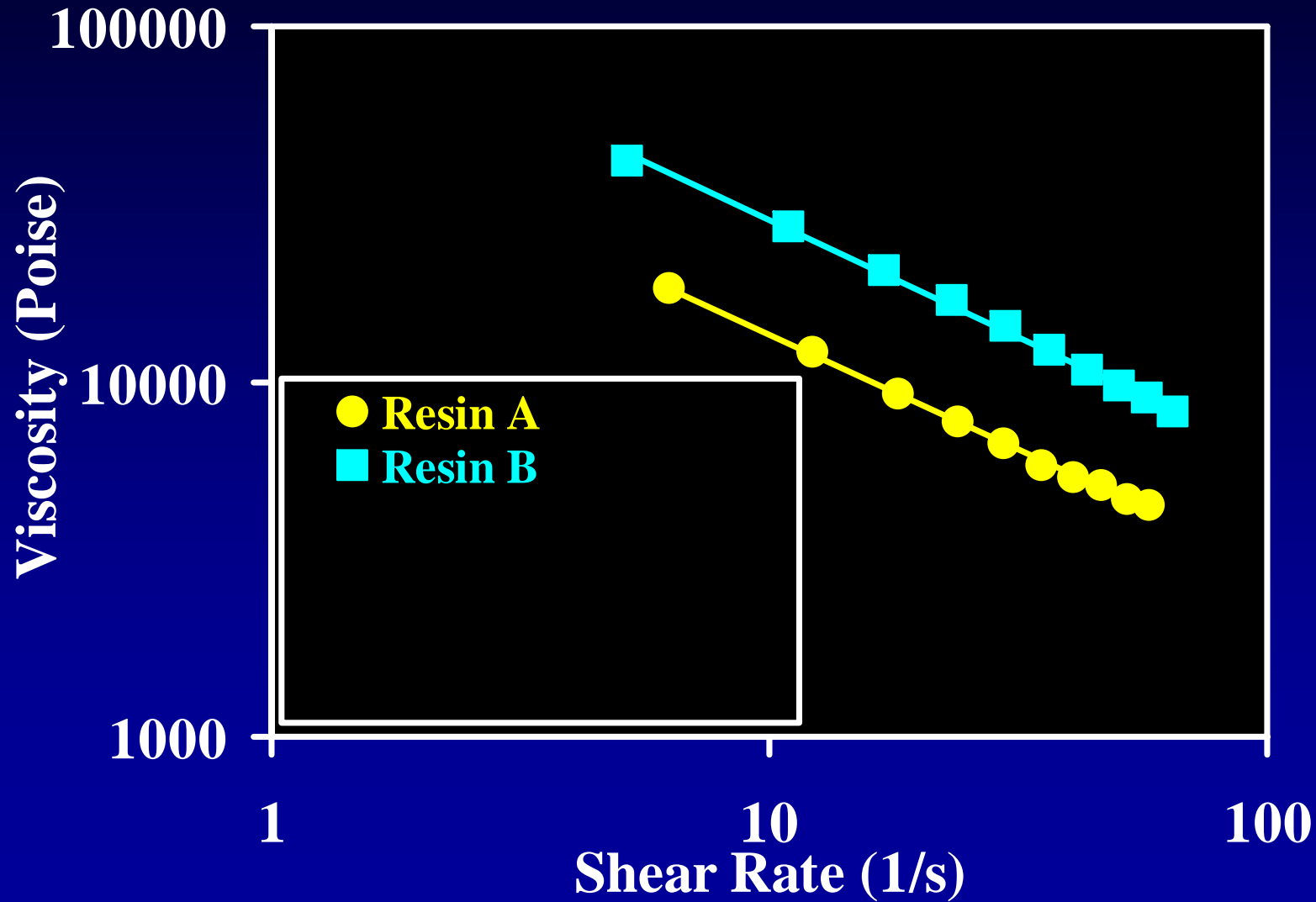
Multilayer Rheology



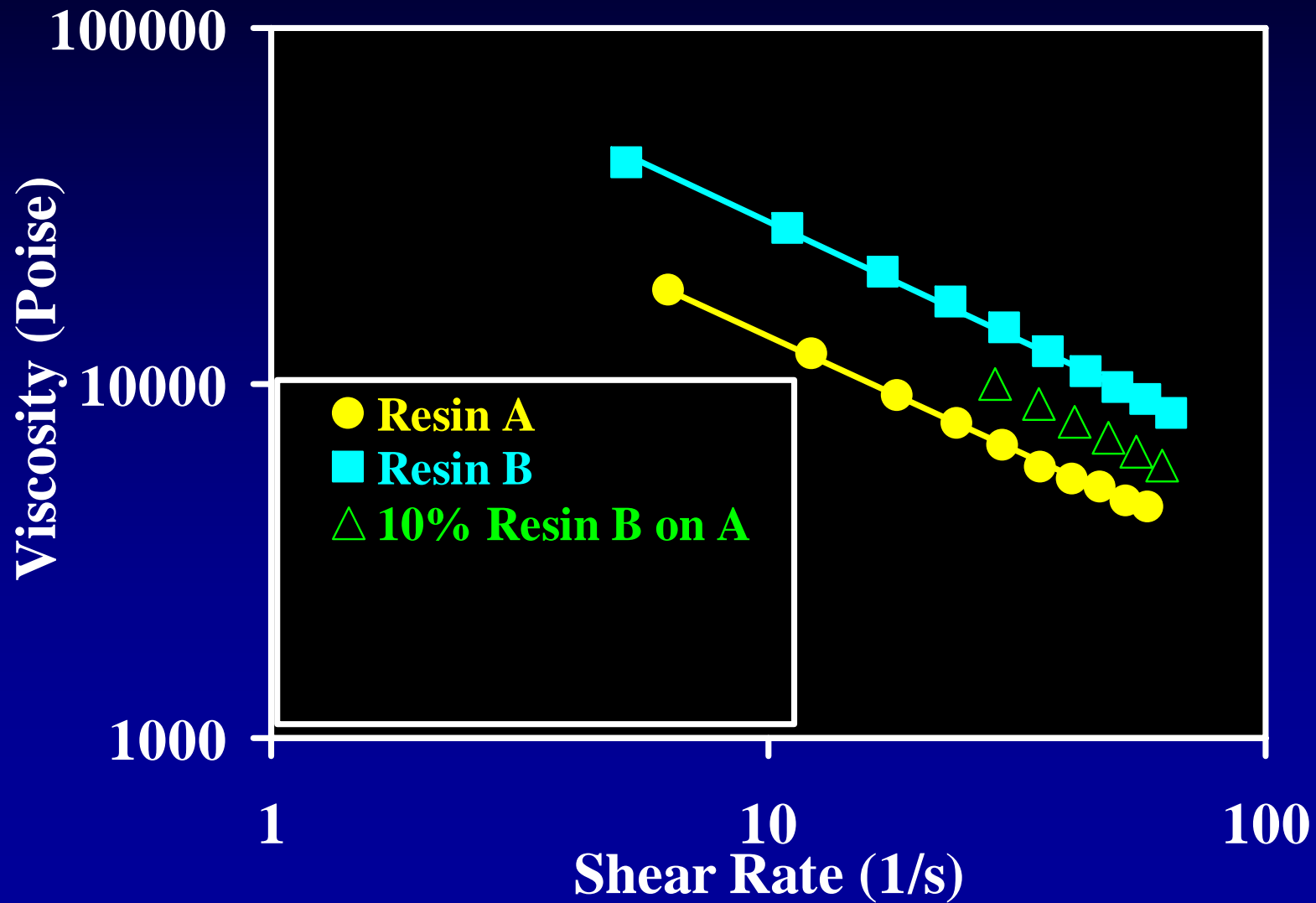
Power Law “K” Function



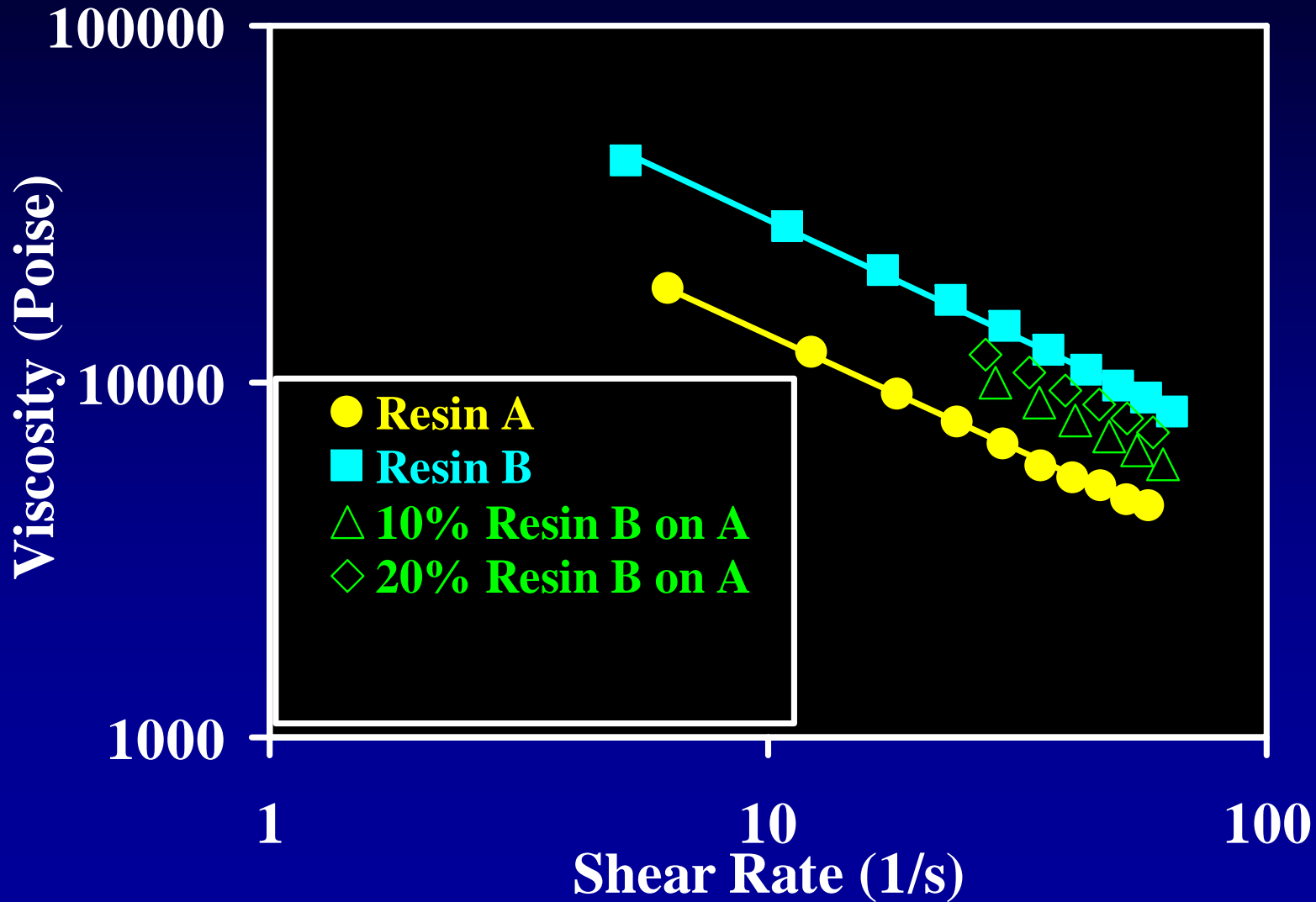
Monolayer Rheology



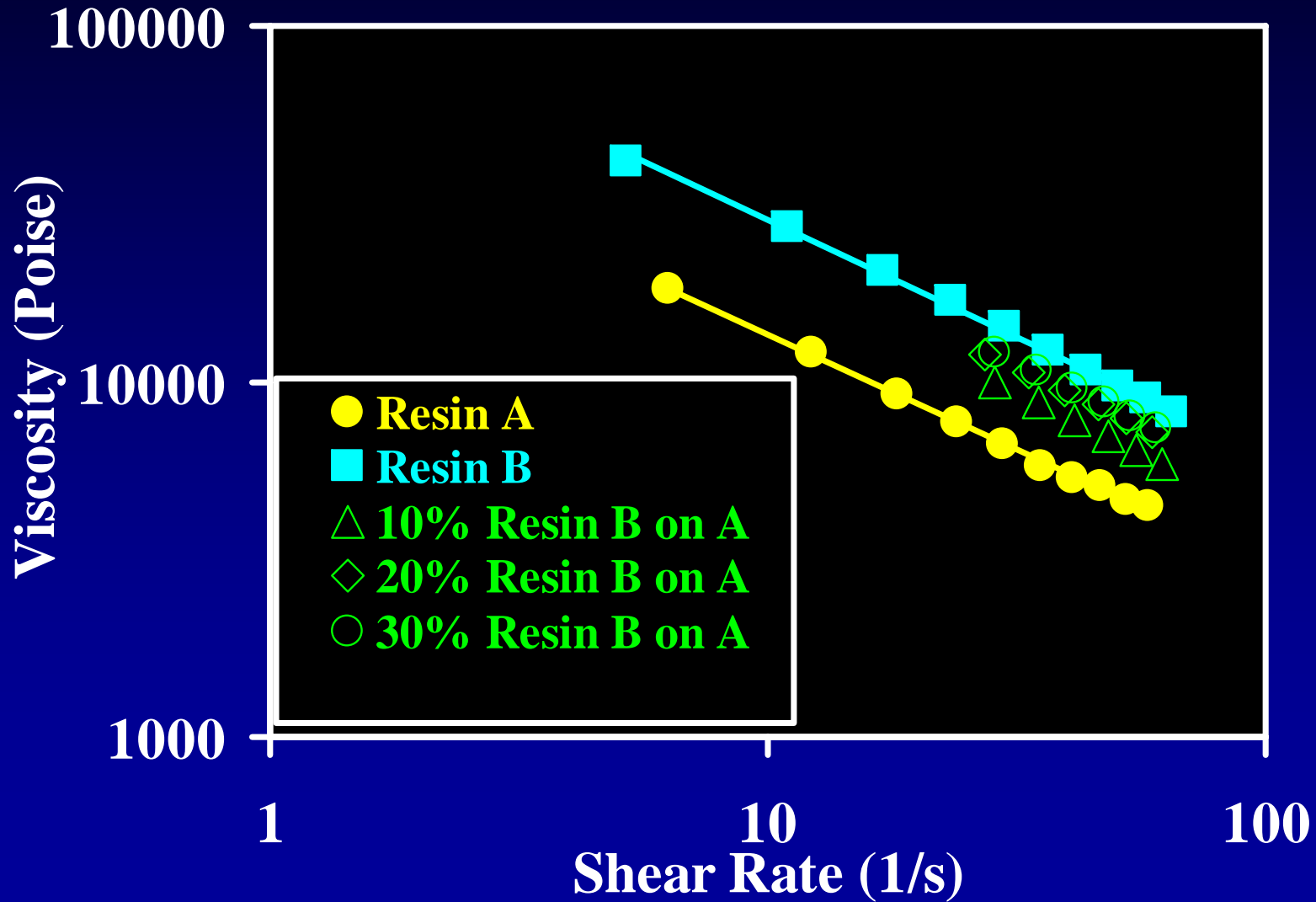
Multilayer Rheology



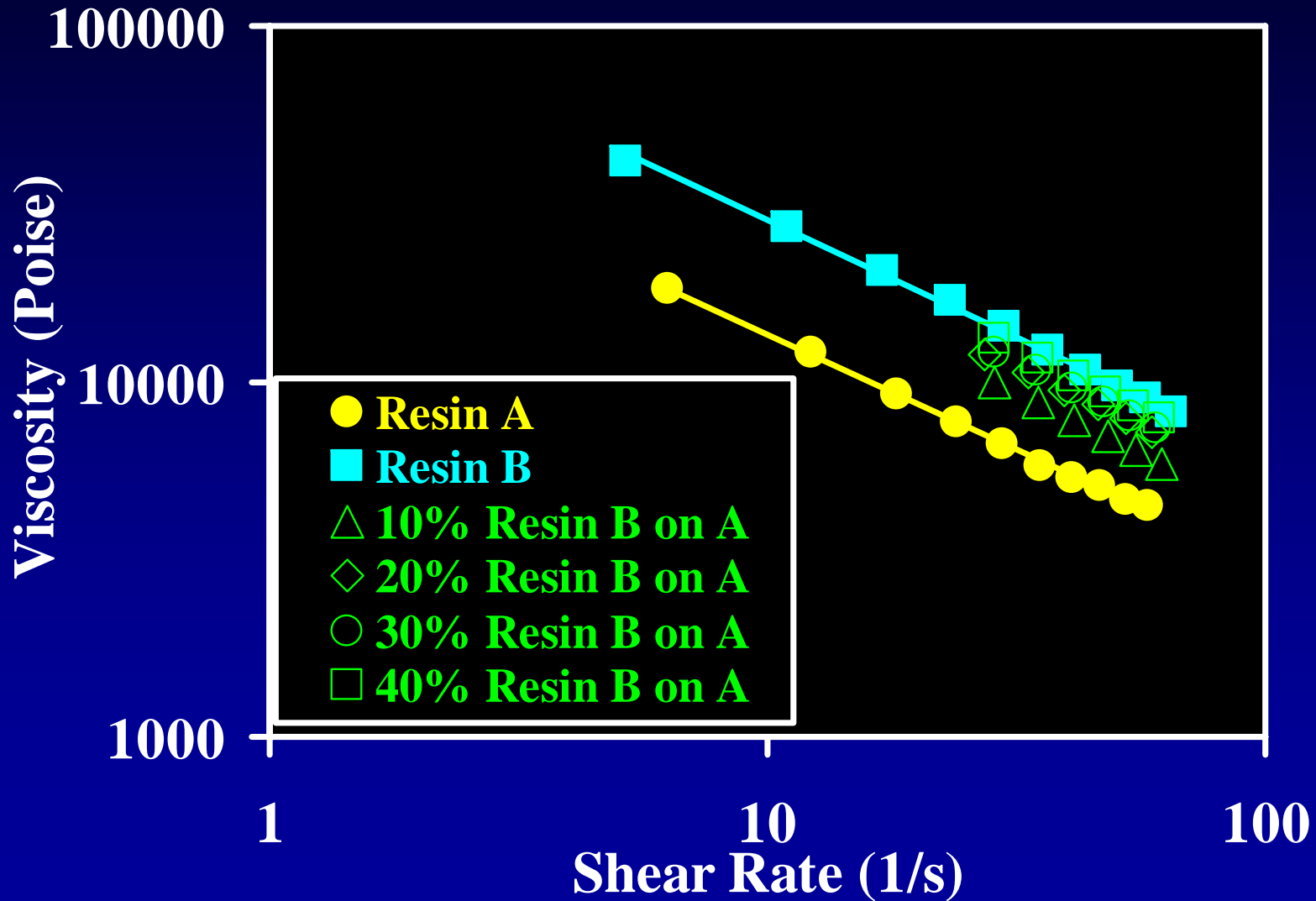
Multilayer Rheology



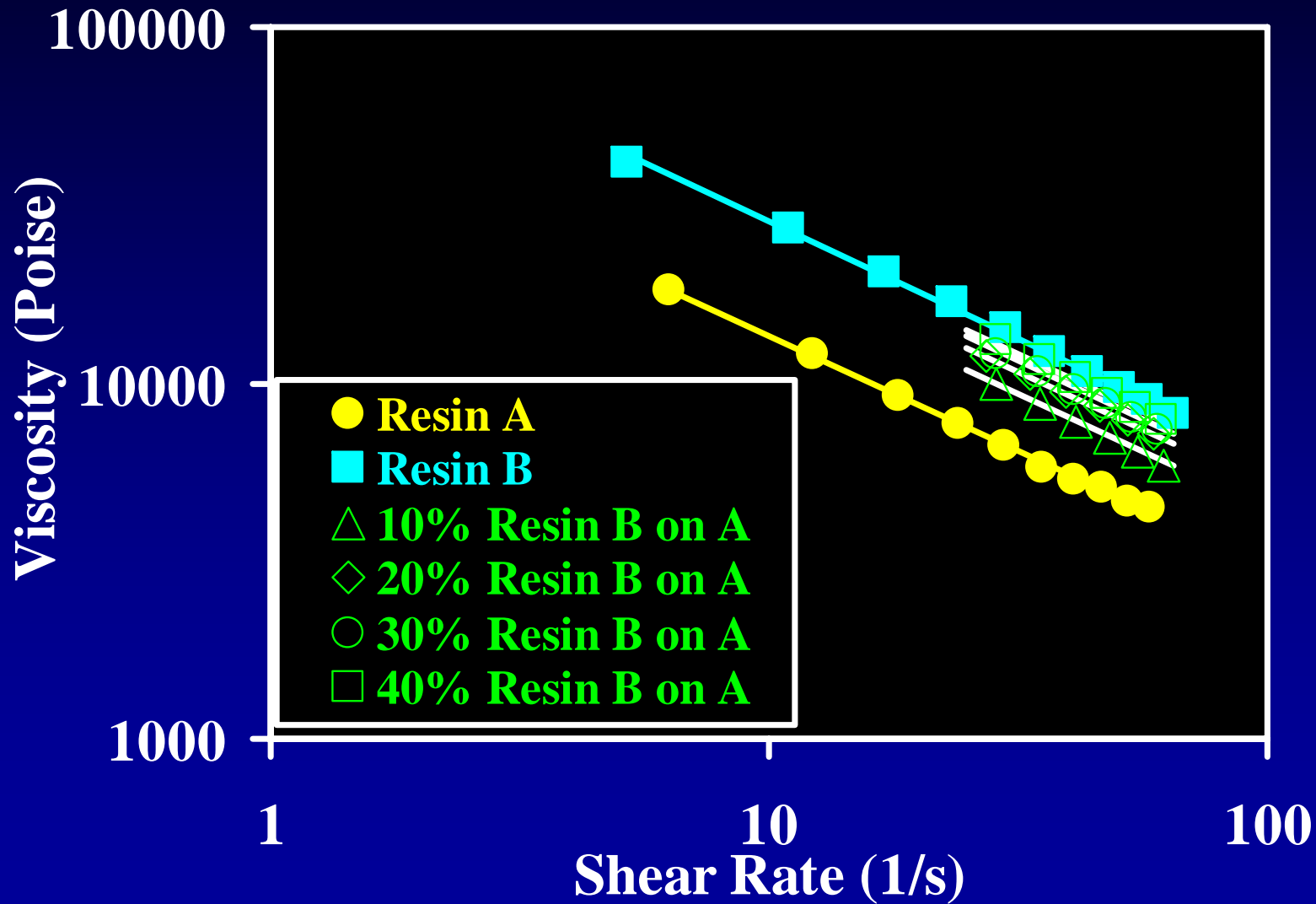
Multilayer Rheology



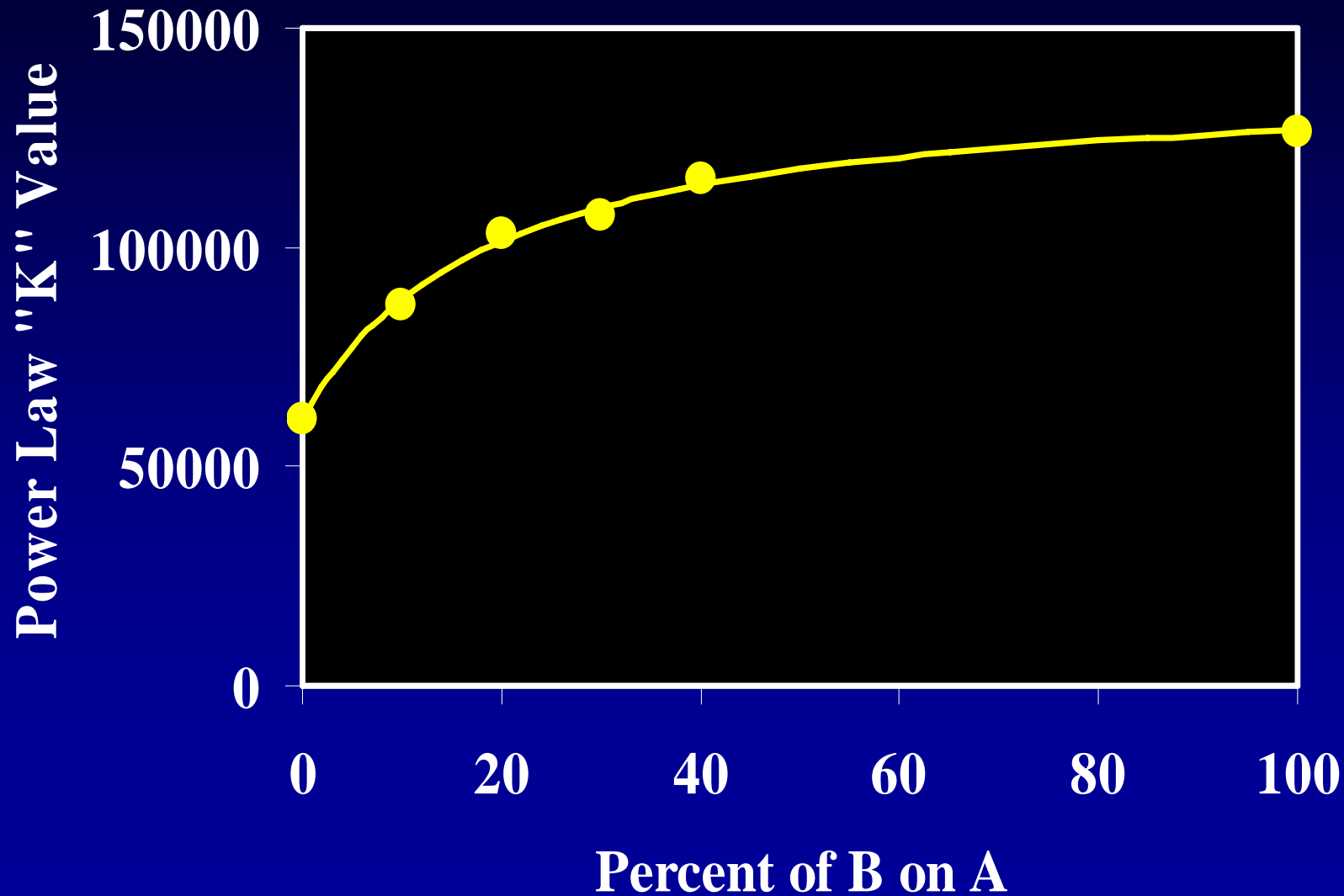
Multilayer Rheology



Multilayer Rheology



Power Law “K” Function

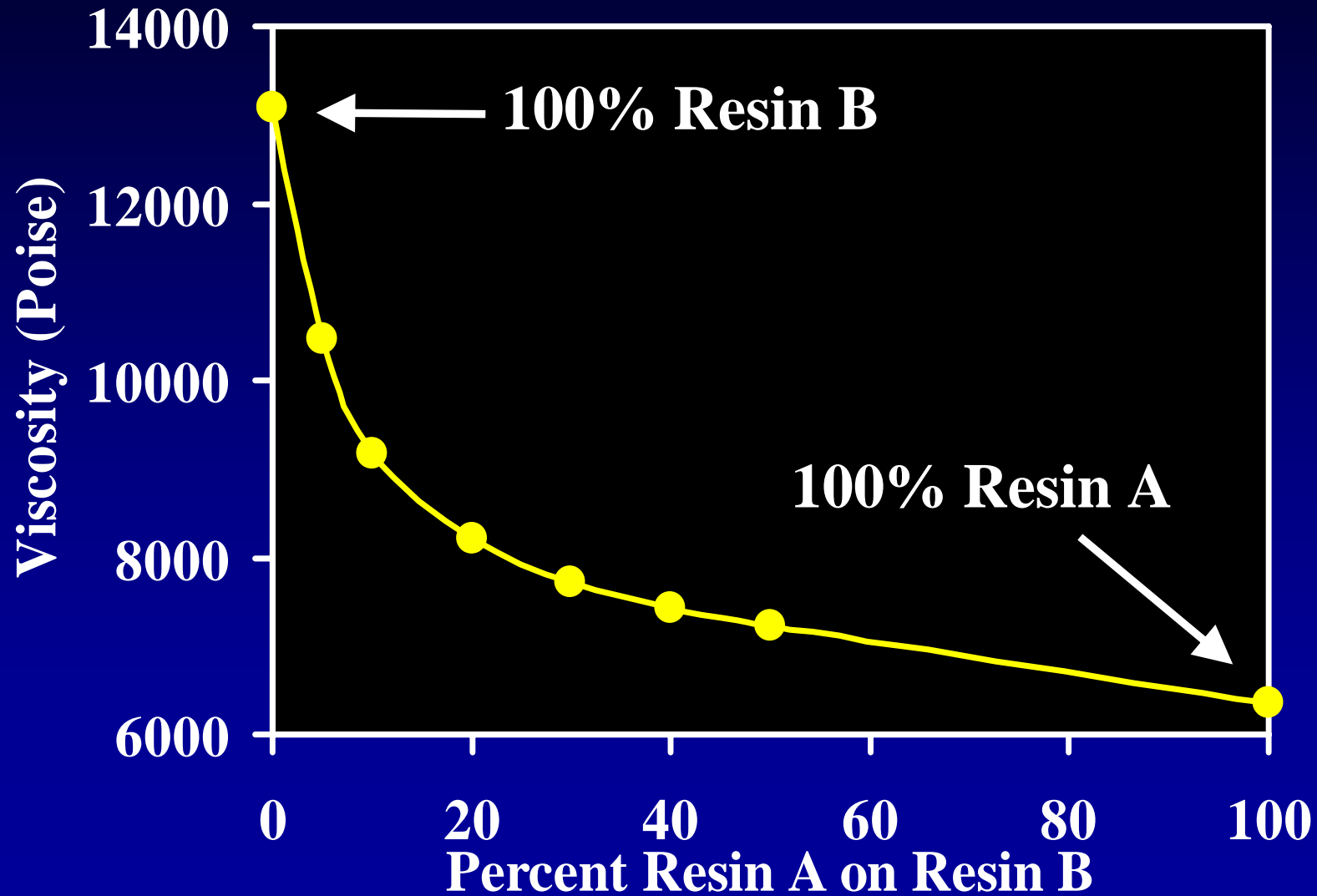


Layer Thickness Effects

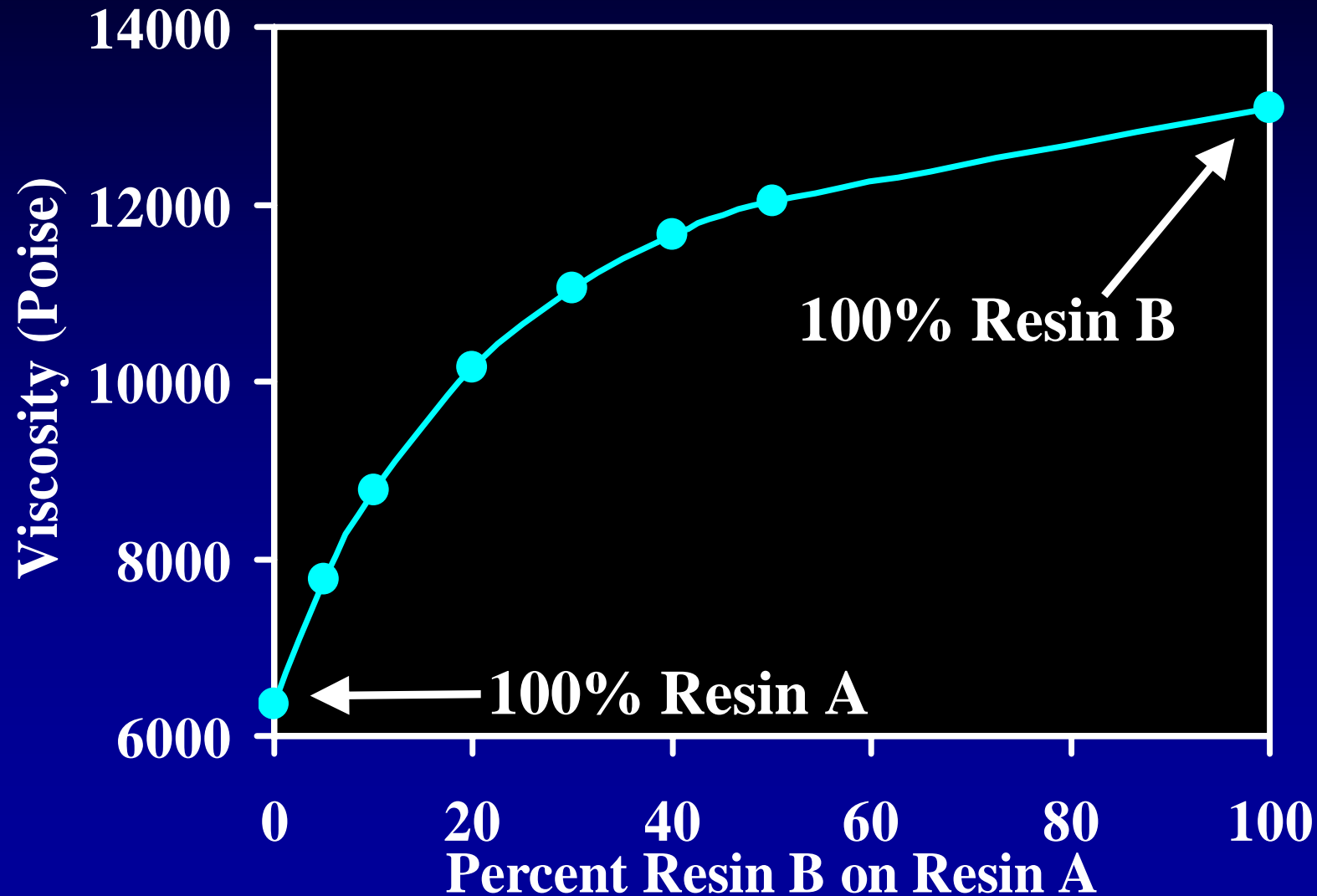
Layer Thickness Effects

- **Measure viscosity of a coextruded structure as a function of skin layer thickness at a constant shear rate**
- **Structures: Resin B encapsulating Resin A and Resin A encapsulating Resin B**
- **Keep total output (shear rate) constant**
- **Vary ratio of skin to core layer**
- **Produce structures with 0, 5, 10, 20, 30, 40, 50 and 100% skin layers**

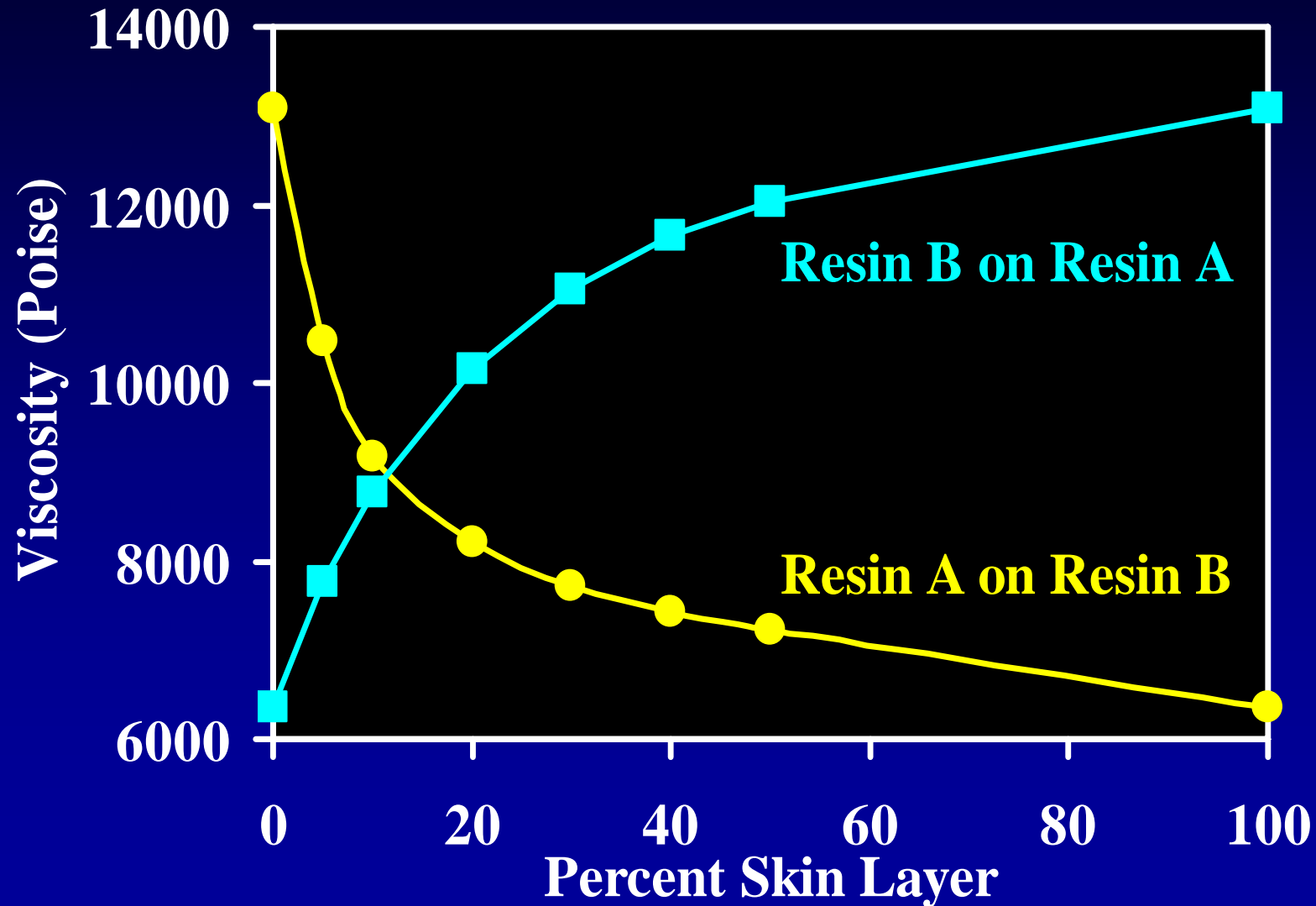
Skin Layer Thickness Effects



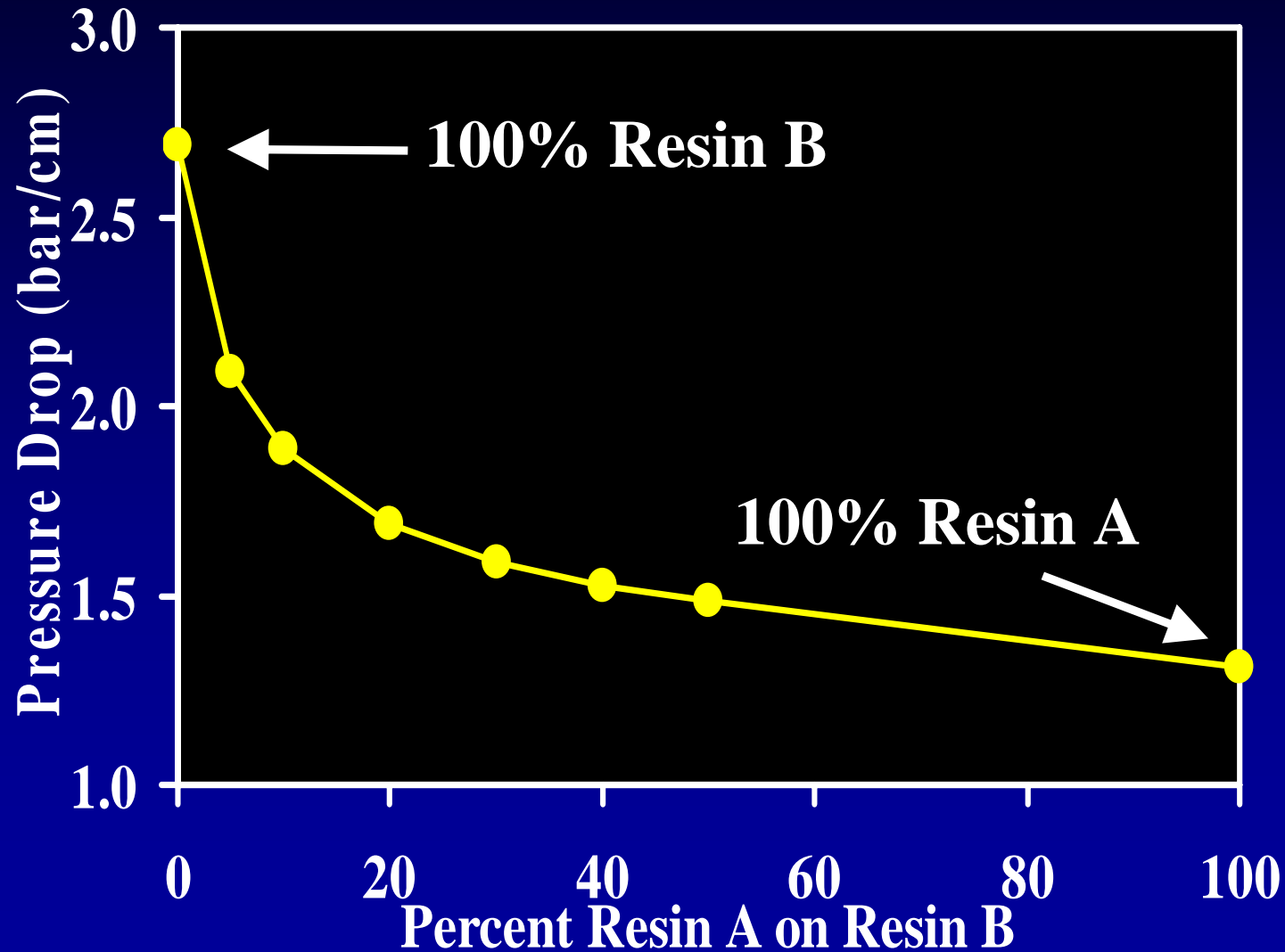
Skin Layer Thickness Effects



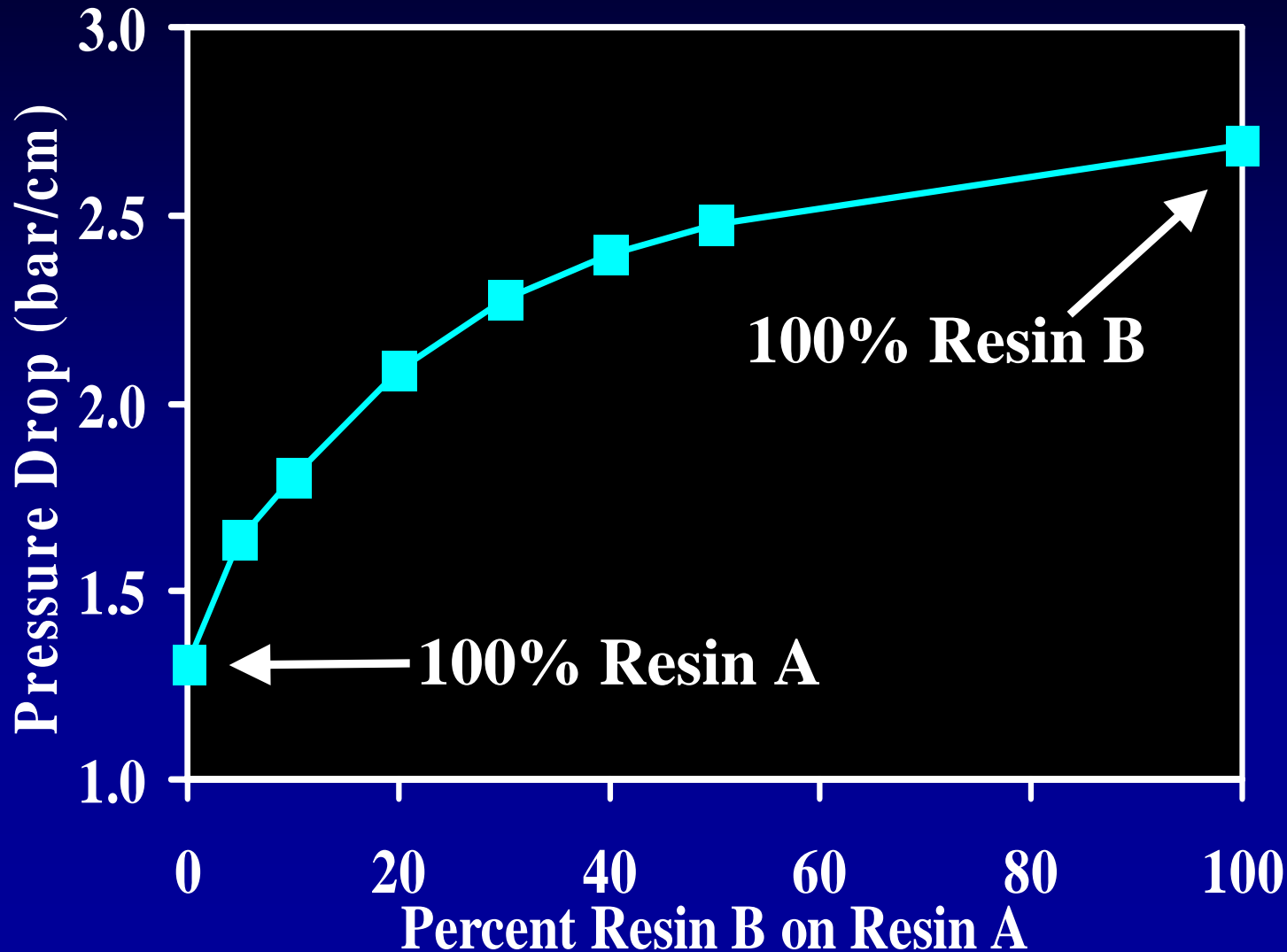
Skin Layer Thickness Effects



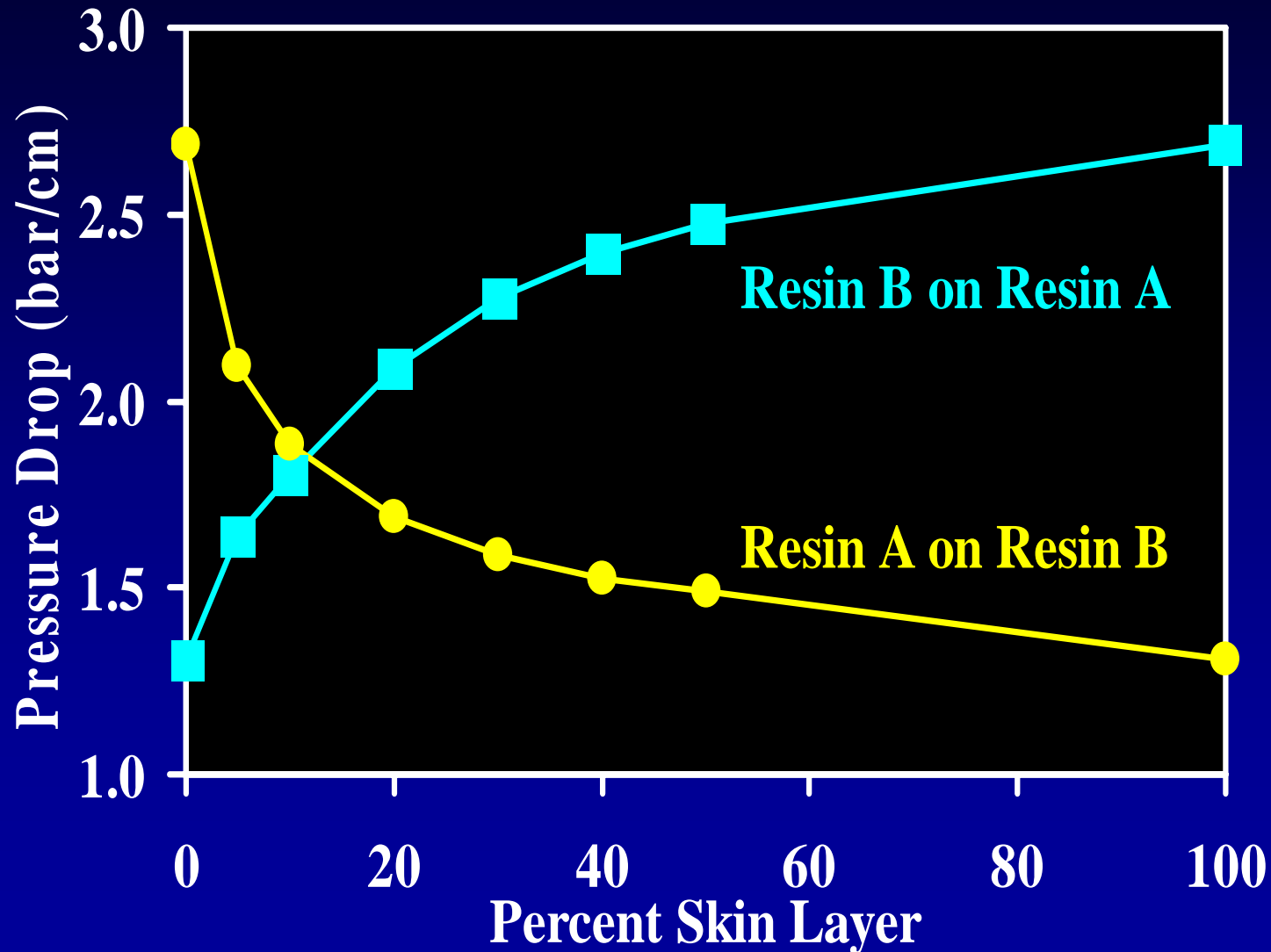
Skin Layer Thickness Effects



Skin Layer Thickness Effects



Skin Layer Thickness Effects



Shear Thinning Effects

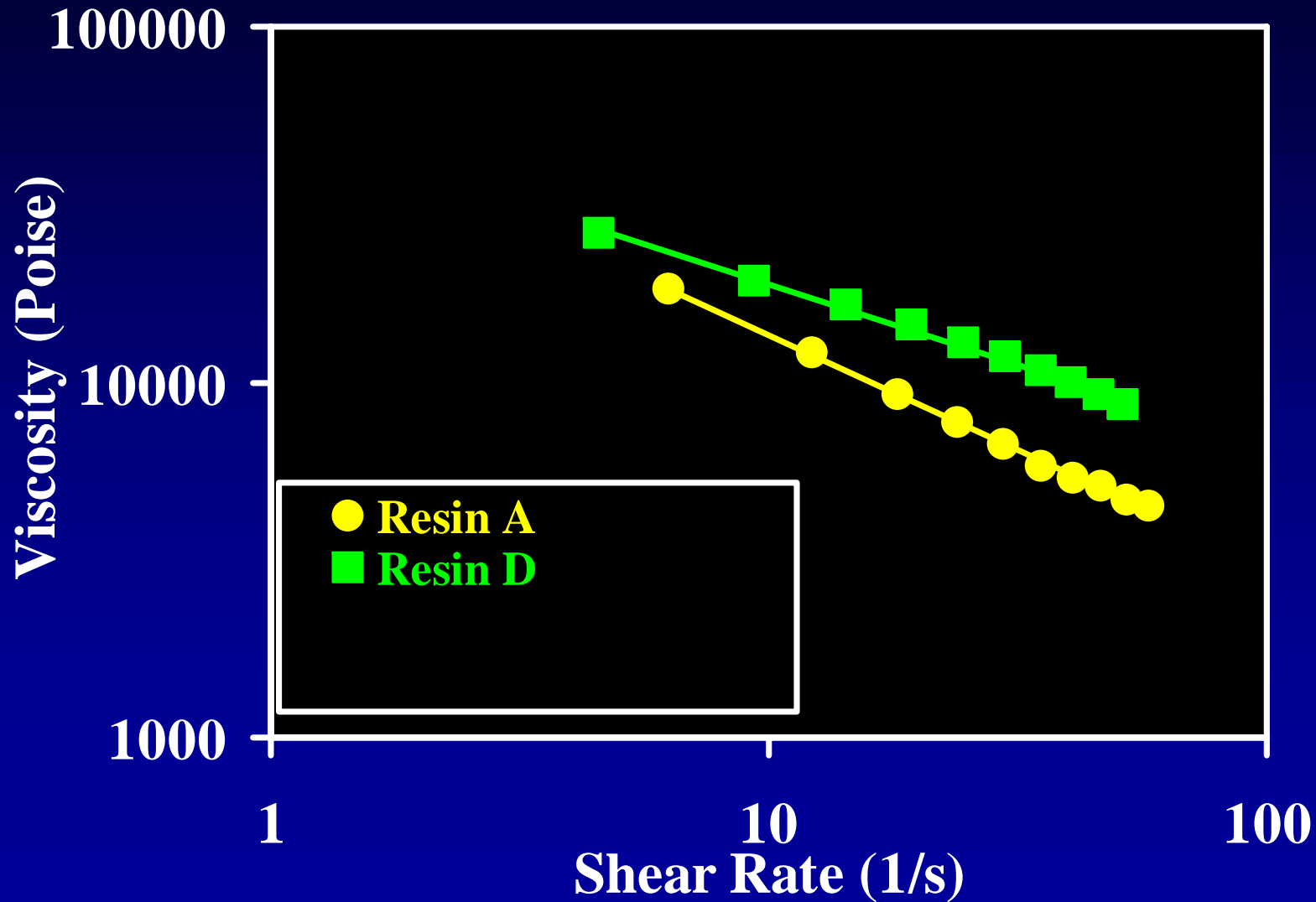
Shear Thinning Effects

- **Measure viscosity of a coextruded structure as a function of shear rate for materials with different shear thinning characteristics**
- **Structure: Resin A encapsulating Resin D and Resin D encapsulating Resin A**
- **Resins have different viscosities**
- **Resins have different shear thinning characteristics**

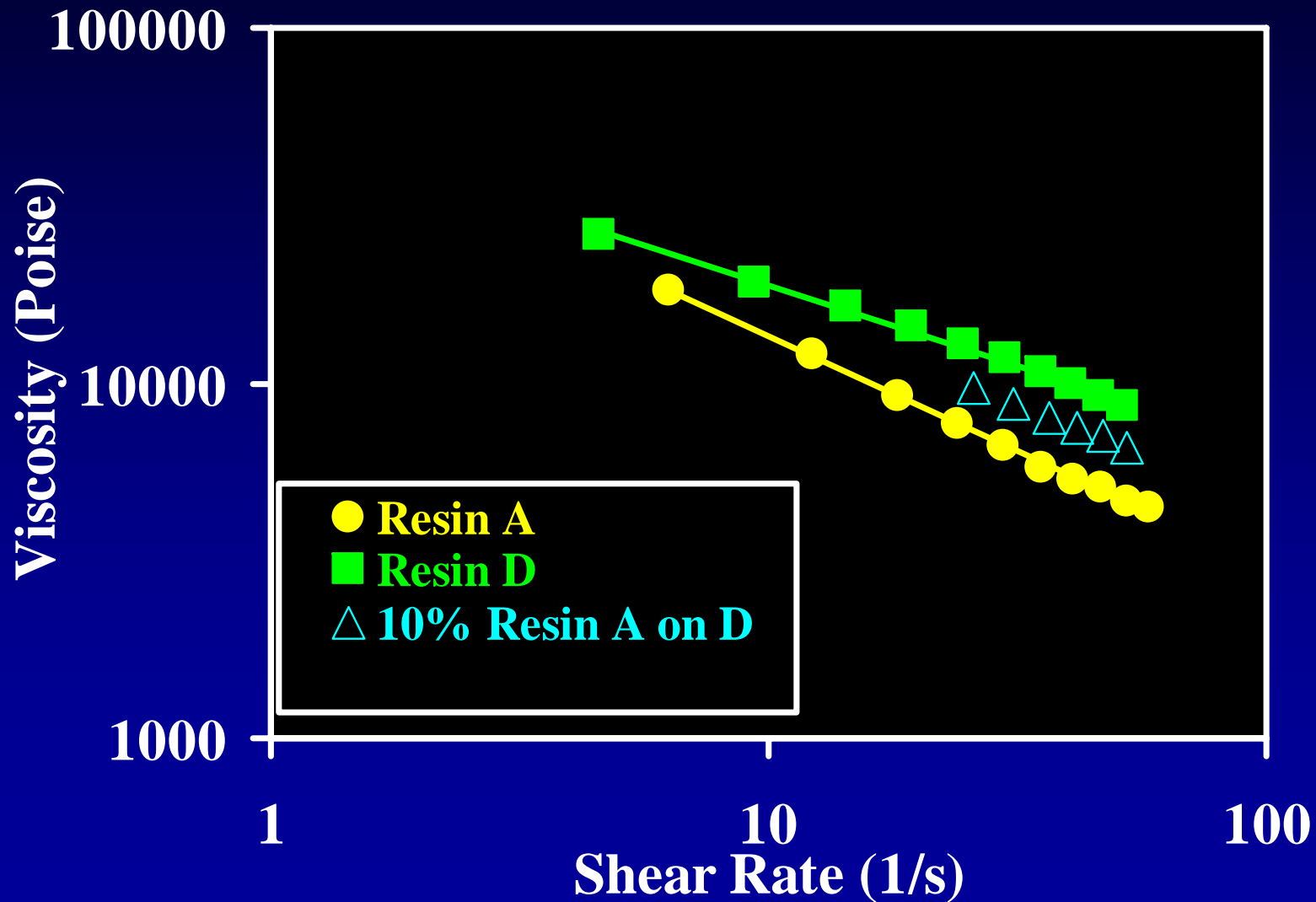
Shear Thinning Behavior

<u>Resin Type</u>	<u>Designation</u>	<u>MI</u>	<u>“n”</u>
Polyethylene	Resin A	8	0.36
Polyethylene	Resin B	2	0.35
Polyethylene	Resin C	1	0.31
Polyethylene	Resin D	5	0.55

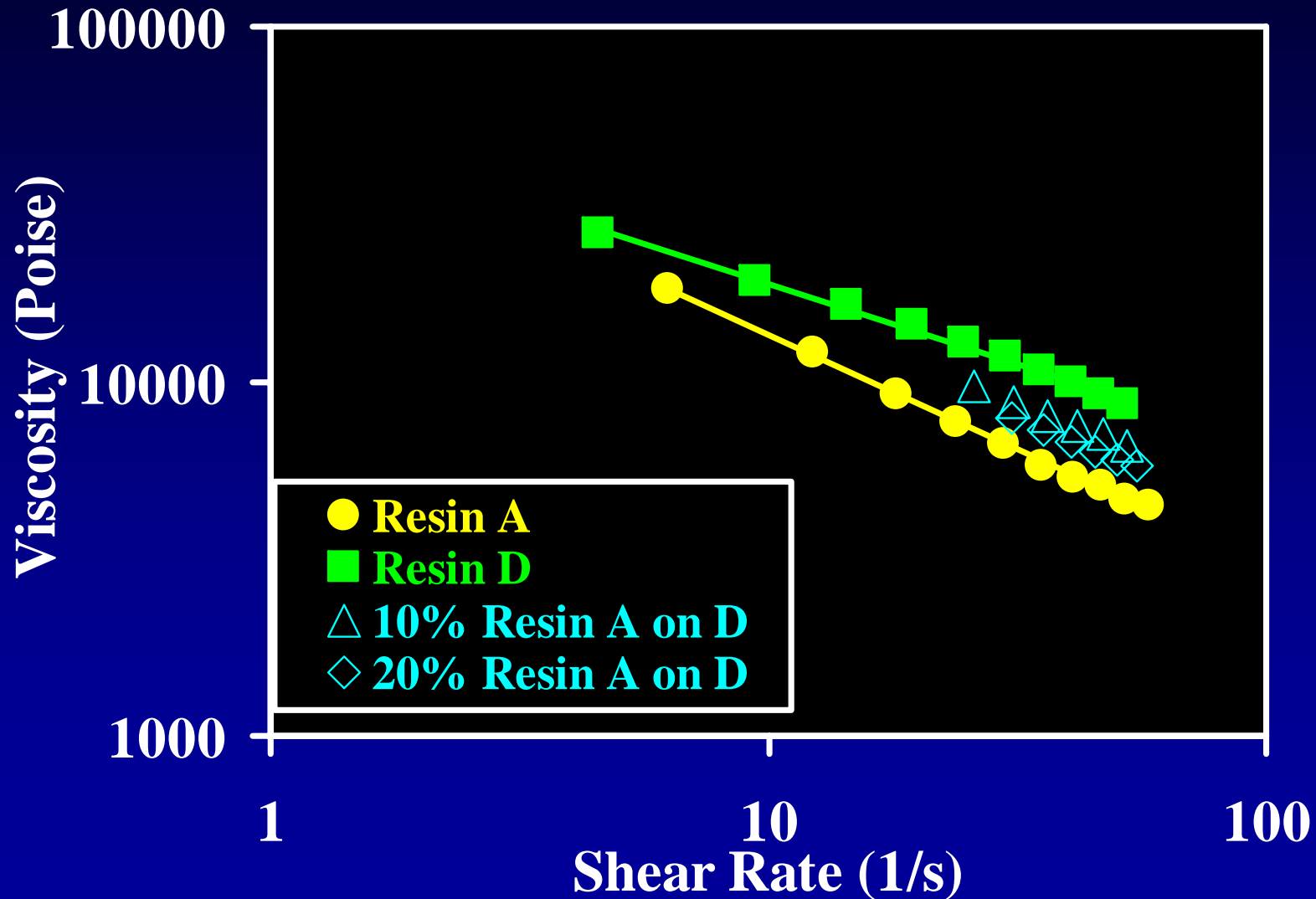
Monolayer Rheology



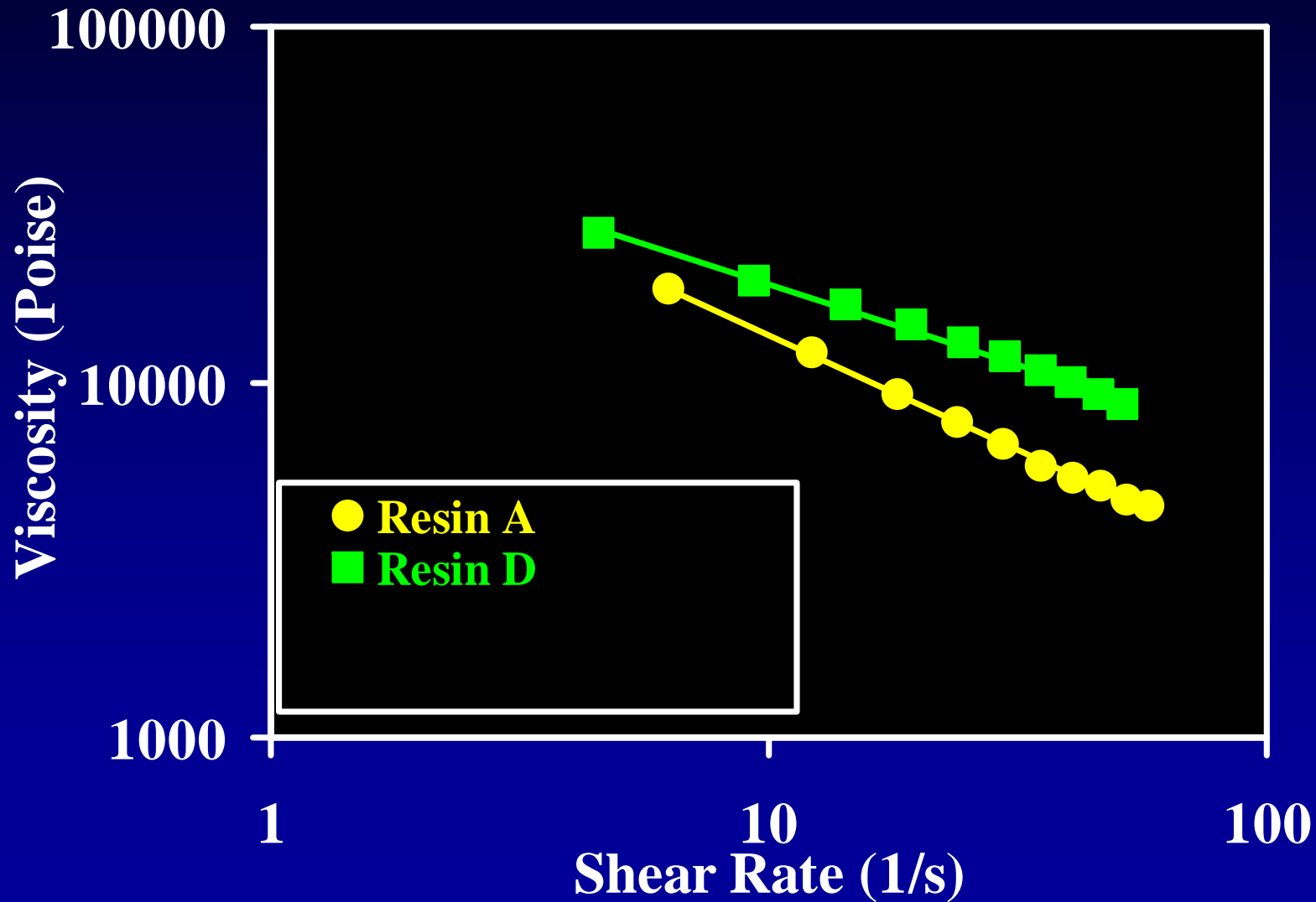
Multilayer Rheology



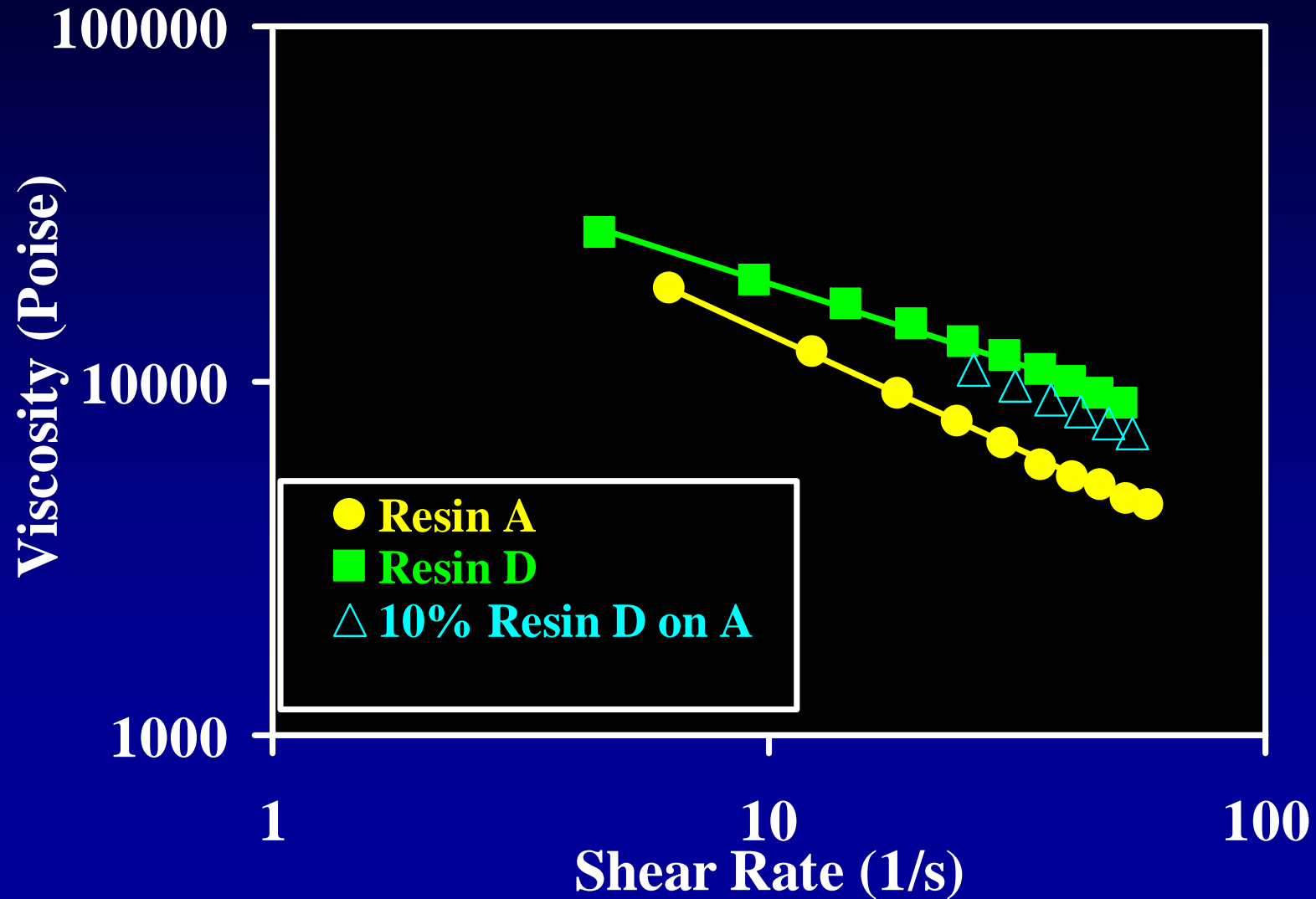
Multilayer Rheology



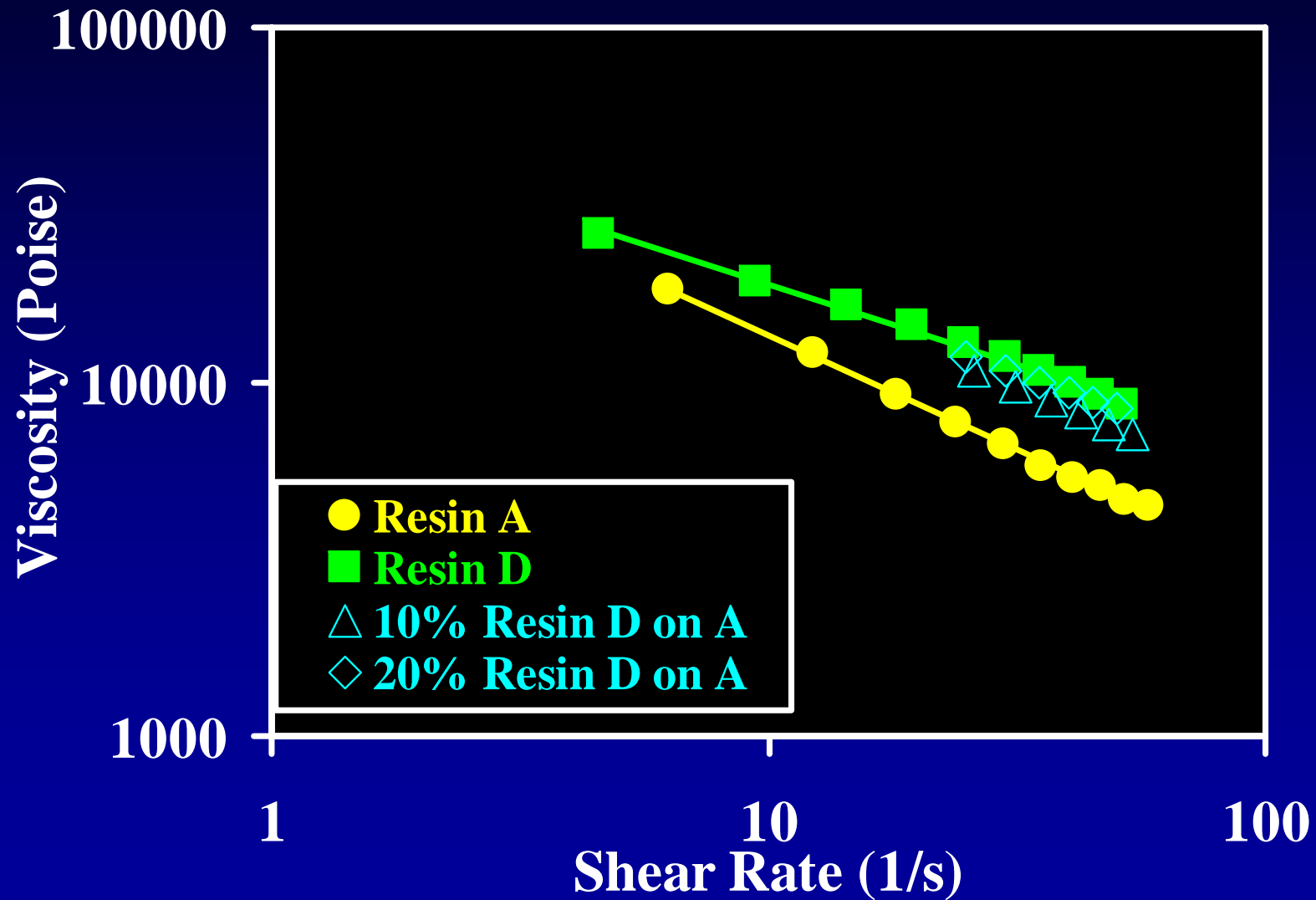
Monolayer Rheology



Multilayer Rheology



Multilayer Rheology



Shear Thinning Behavior

<u>Structure</u>	<u>Power Law Index “n”</u>
Resin A	0.36
20% A on D	0.40
10% A on D	0.46
Resin D	0.55

Shear Thinning Behavior

<u>Structure</u>	<u>Power Law Index “n”</u>
Resin A	0.36
10% D on A	0.44
20% D on A	0.51
Resin D	0.55

Summary

- **A unique apparatus has been developed to measure the rheology of coextruded structures**
- **A series of experiments were run to evaluate the effects on processability of the rheology of a coextruded structure by varying the:**
 - **layer viscosity**
 - **layer thickness**
 - **layer placement**
 - **shear thinning behavior**

Summary

- **These experiments showed that the rheology of a coextruded structure is a complex function the viscosity, thickness, placement, and shear thinning behavior of the individual layers**
- **These results should help with determining the processability of coextruded structures based on the pressure drop in the process**

Thank You

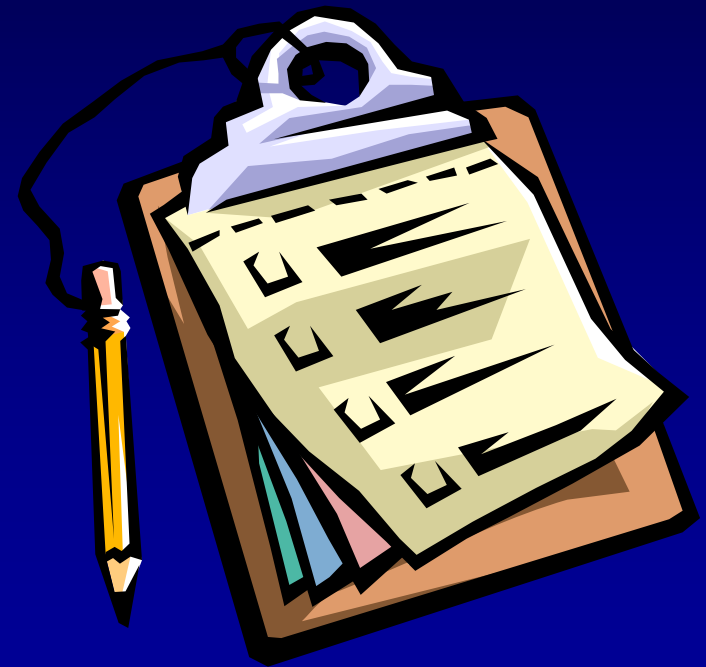
PRESENTED BY

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*Please remember to turn in
your evaluation sheet...*