Material Strength in Polymer Shape Deposition Manufacturing

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Abstract

Shape Deposition Manufacturing (SDM) is a layered manufacturing process involving an iterative combination of material addition and material removal. Polymer SDM processes have used castable thermoset resins to build a variety of parts. The strength of such parts is determined by the bulk material properties of the part materials and by their interlayer adhesion. This paper describes tensile testing of three thermoset resins used for SDM - two polyurethane resins and one epoxy resin. Both monolithic specimens and specimens with two interlayer interfaces were tested. Interlayer tensile strengths were found to vary greatly among the three materials, from 5-40 MPa.

Introduction

Shape Deposition Manufacturing (SDM) is a layered manufacturing process involving an iterative combination of material addition and material removal [1] [2]. Two materials are used: a part material and a sacrificial support material which surrounds it. Objects are built in layers by depositing and machining part material. Layers containing under-cut features are produced by replicating them from complementary features machined into surrounding support material. When objects are completely finished, they are freed by dissolving the support material. The current polymer part materials are thermoset resins, and the current support materials are machinable waxes although other combinations have been used in the past [3]. The part materials are two-part, castable resins which must be mixed together and immediately cast onto the growing part. Vacuum degassing is used to prevent voids caused by air bubbles. The mixing and casting process is currently performed manually, but its automation would be straightforward using equipment similar to that developed for the Mold SDM process [4]. Polymer SDM is especially suited for building parts which are relatively large but have few transitions between non-undercut and under-cut features. Such parts can be made using a few thick layers at a reasonable process speed.

Since Shape Deposition Manufacturing is a layered manufacturing process, the mechanical properties of SDM parts are dependent both upon the bulk material properties and upon the quality of the resulting interlayer bonds. The successive deposition of cast layers also produces anisotropic mechanical properties in polymer SDM parts; these properties are investigated in the present study.

Polymer SDM Materials

Several materials are currently used for the production of polymer parts via SDM. All of the part materials are castable thermoset polymers while the support material is a machinable wax. The first part material is LUC 4180 polyurethane from Adtech Plastic Systems Corp. of Charlotte, Michigan. This material is reasonable strong, has good impact properties, and can be machined about twelve hours after casting. It was initially chosen for its low water absorption for use in wearable computers for divers [5]. However, later experiments indicated that this material suffers from poor interlayer bonding. The second material is TDT 205-3 polyurethane from Ciba Specialty Chemicals Corp. in East Lansing, Michigan. This material was chosen for its rapid cure speed; successive layers can be machined two hours after casting. Finally, the third material is EE-501/530 epoxy, also from Adtech Plastic Systems Corp. This material is a highly filled encapsulation resin which was chosen for its good adhesion to other materials; it was hoped that it would also adhere well to previous layers of the same material. This material requires a significantly longer cure time than either of the other materials; twenty-four hours' delay is required between casting and machining. Manufacturer's specifications for each of the materials are summarized in Table 1.

Material	Adtech LUC 4180 Polyurethane	Ciba TDT 205-3 Polyurethane	Adtech EE-501/530 Epoxy
Ultimate Tensile Strength (MPa)	55	23	42
Elongation to Failure	15%	9%	1%
Hardness (Shore D Scale)	78-80	70	86-89
Mixed Viscosity (cPs)	800-900	80	3500
Reference	[6]	[7]	[8]
Cure Time before Machining	12 hours	2 hours	24 hours

Table 1. Manufacturer's Specifications

Tensile Strength Testing

The objective of these tests is to determine the tensile strengths of parts made via SDM with these three materials. Since SDM is a layered manufacturing process, test specimens constructed in different orientations may have different tensile strengths. This difference might be particularly pronounced when comparing test specimens built within a single layer to those which extend over multiple layers, since interlayer bonds may be weaker than the bulk material. ASTM D 638-96 specifies shapes and sizes for plastic tensile specimens, as well as conditioning and testing procedures [9]. This standard describes methods of measuring material properties, and was used as a baseline for the present study. The recommended dimensions for ASTM Type I dogbone tensile specimens were used: a 13.0 mm x 57.0 mm narrow section of 3.2 mm thickness, with 20mm-wide grip ends and a total length of 170 mm (as shown in Figure 1).

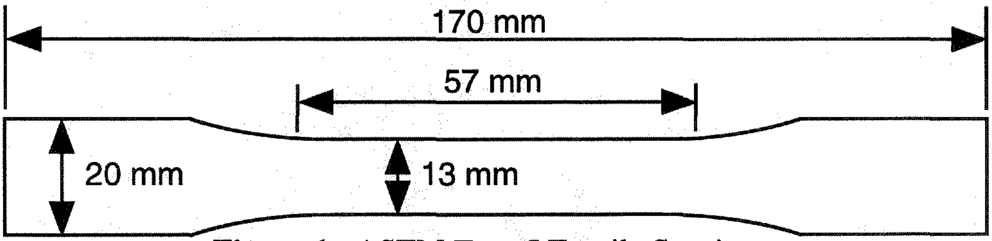

Figure 1. ASTM Type I Tensile Specimen

Ideally, bulk and interlayer tensile strengths would be compared by building standard ASTM tensile specimens both perpendicular and parallel to the build direction. The specimens built perpendicular to the build direction would be planar and monolithic, built horizontally within a single layer of material. The specimens built parallel to the build direction would be vertical, composed of many layers of material. Both ideal specimens are shown in Figure 2.

Building thin-layer vertical tensile specimens, however, would take a long time since single layers of one of these materials require 24-hour cure times. Therefore, a simplified dual-interface test specimen was developed which can be made in two cure cycles; this design is shown in Figure 3. These specimens are constructed by casting a complete layer of material, machining a transverse trench through the layer, casting material into that trench, and finally machining the outlines of individual tensile specimens. The resulting specimens have two interlayer interfaces, rather than the *n-1* interfaces present in an *n*-layer vertical tensile specimen. The tensile strengths of these dual-interface specimens will be expected to represent an upper bound on the tensile strength of an actual vertical *n*-layer tensile specimen made via SDM.

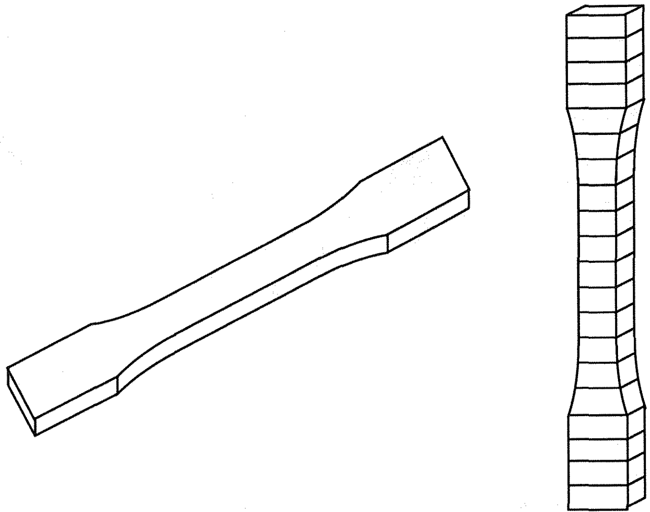

Figure 2. Ideal Planar and Vertical Tensile Specimens

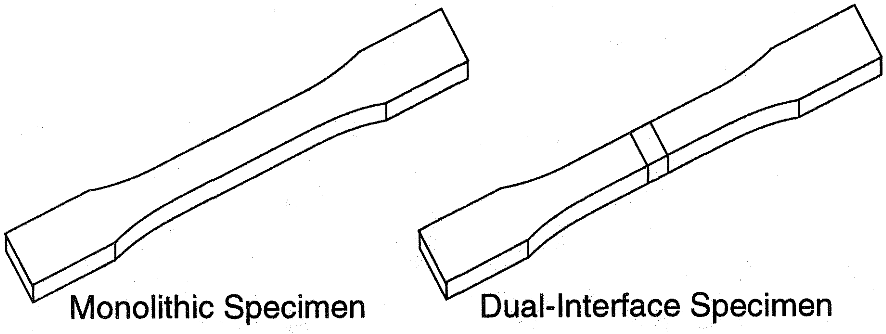

Figure 3. Monolithic and Simplified Dual-Interface Tensile Specimens

The tensile tests for LUC 4180 were conducted in four rounds, split between General Motors and Stanford University. Several different batches of LUC 4180 were used, including both material from newly opened containers and older material from containers which had been opened before. While this may result in variations in material properties, it is consistent with part production methods. The TDT 205-3 and 501/530 material used for tensile specimens came from newly opened containers.

After the initial casting, the monolithic tensile specimens were cut out, and trenches were cut into the dual-interface specimens. Additional polymer material was then cast into the trenches and allowed to cure. The dual-interface specimens were cut out, and all specimens were aged for two weeks to ensure complete curing. LUC 4180 specimens were machined 18-26 hours after casting, except for those of Series II (which were machined four days after casting). The TDT 205-3 polyurethane specimens were machined between 4-6 hours after casting, and the 501/530 epoxy specimens were machined between 24-46 hours after casting. As would be expected during part production, very little surface preparation was performed between the initial machining step and the second casting step. Chips and any residue from the machining process were cleared with compressed air, and the surfaces were wiped with Kimwipes; no cleaning solvents were used.

Since the objective was to determine the strengths of parts made through SDM, several specimen preparation steps normally recommended by the ASTM were skipped because they might overestimate actual strengths. The D 638 standard specifies smoothly polished surfaces, but the machined or replicated surfaces resulting from the SDM process were tested without any additional polishing. On dual-interface tensile specimens, there were slight thickness variations between the initial casting and the second casting. These artifacts were left undisturbed rather than attempting to remove them.

Series I of the LUC 4180 tests was conducted at GM, while the remaining series were conducted at Stanford. All of the tests of TDT 205-3 and 501/530 were conducted at Stanford. The tests at GM were performed on an Instron machine with an extensometer, while the Stanford tests were performed on an Instron machine without an extensometer. The Series I LUC 4180 tests were conducted at a constant crosshead speed of 50 mm/min, while all other tests were conducted at 0.2"/min (5.1 mm/min). At Stanford, data from LUC 4180 Series II and III tests was collected using a chart recorder while Series IV, TDT 205-3, and 501/530 data was digitized and stored by a Labview program.

Results

The tensile testing produced some interesting results for both monolithic and dual-interface specimens. The monolithic LUC 4180 polyurethane specimens failed very differently from monolithic specimens of the other materials. LUC 4180 specimens yielded and formed necks before finally breaking; both TDT 205-3 polyurethane and 501/530 epoxy failed in a more brittle fashion. A summary of the test data is presented in Table 2, and typical plots of applied stress versus crosshead position for monolithic specimens are shown in Figure 4. LUC 4180 failed at the highest stresses, followed by 501/530, while TDT 205-3 failed at much lower stresses.

Dual-interface specimens produced more surprising results. There were great difficulties manufacturing LUC 4180 specimens with dual interfaces; 8 out of 18 were destroyed during removal from their support material substrates. The LUC 4180 specimens always failed exactly at the interfaces, at very low stresses. The handling difficulties indicate that the average tensile strength would have been even lower if all LUC 4180 specimens had been successfully tested. The TDT 205-3 polyurethane specimens, on the other hand, always failed away from the dual interfaces, and failed at nearly the same stresses as the monolithic specimens had. The 501/530 epoxy specimens usually failed very close to or at the dual interfaces, and also failed at a large fraction of their monolithic failure stresses. Typical plots of applied stress versus crosshead position for the dual-interface specimens are shown in Figure 5. Overall, the 501/530 epoxy dual-interface specimens failed at the highest stresses, while the TDT 205-3 specimens failed at half those stresses, and LUC 4180 specimens failed below one eighth of the 501/530 values. A plot of all test data is shown in Figure 6.

	LUC 4180	TDT 205-3	501/530
Monolithic Specimens	58 MPa	22 MPa	49 MPa
Dual-Interface Specimens	4.6 MPa	20 MPa	40 MPa

Monolithic LUC 4180 specimens: engineering stress at yield
All other specimens: engineering stress at break

Table 2. Average Tensile Strengths

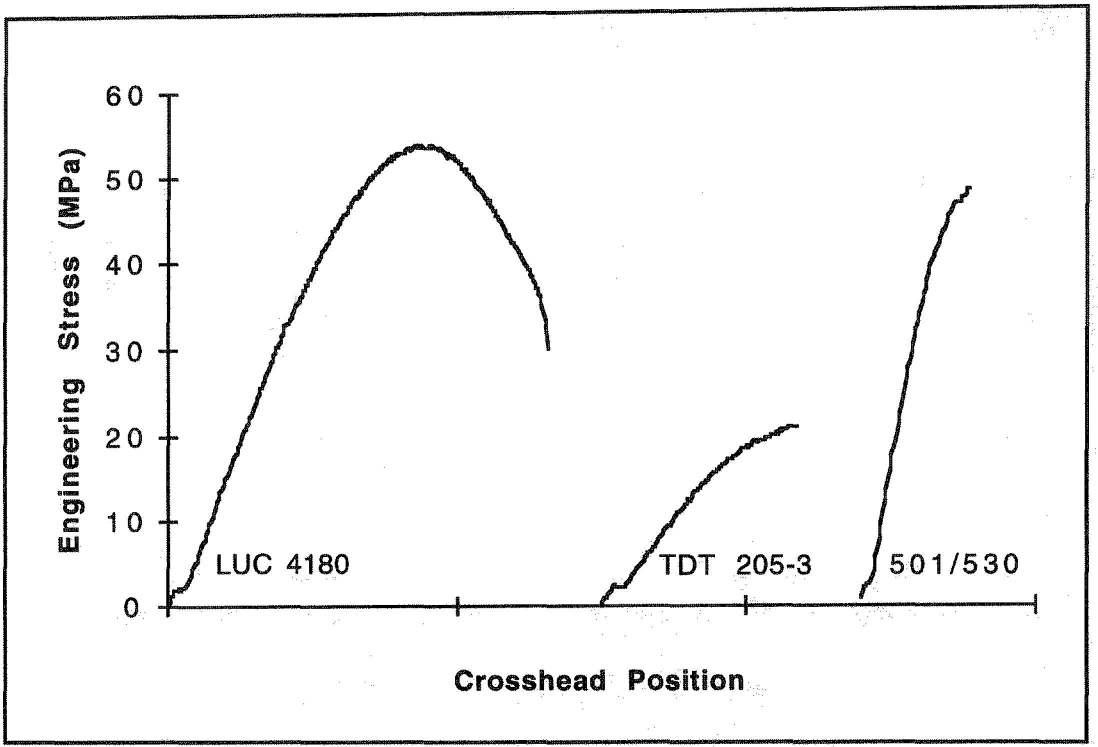

Figure 4. Typical Tensile Tests of Monolithic Specimens

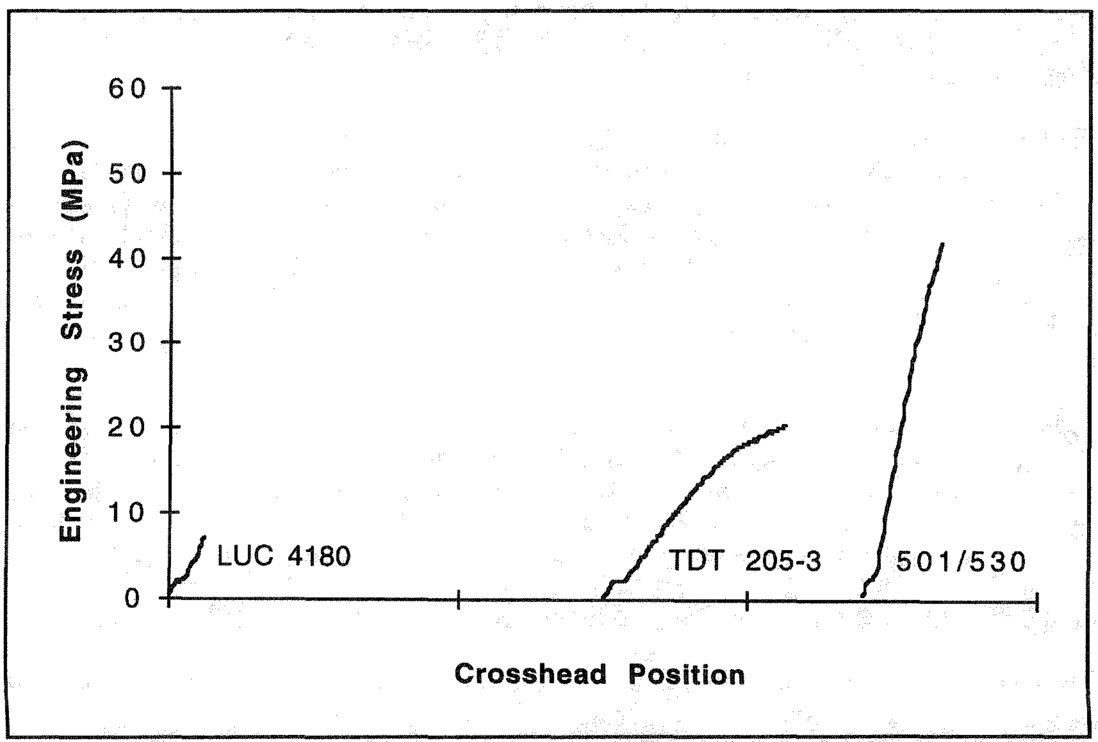

Figure 5. Typical Tensile Tests of Dual-Interface Specimens

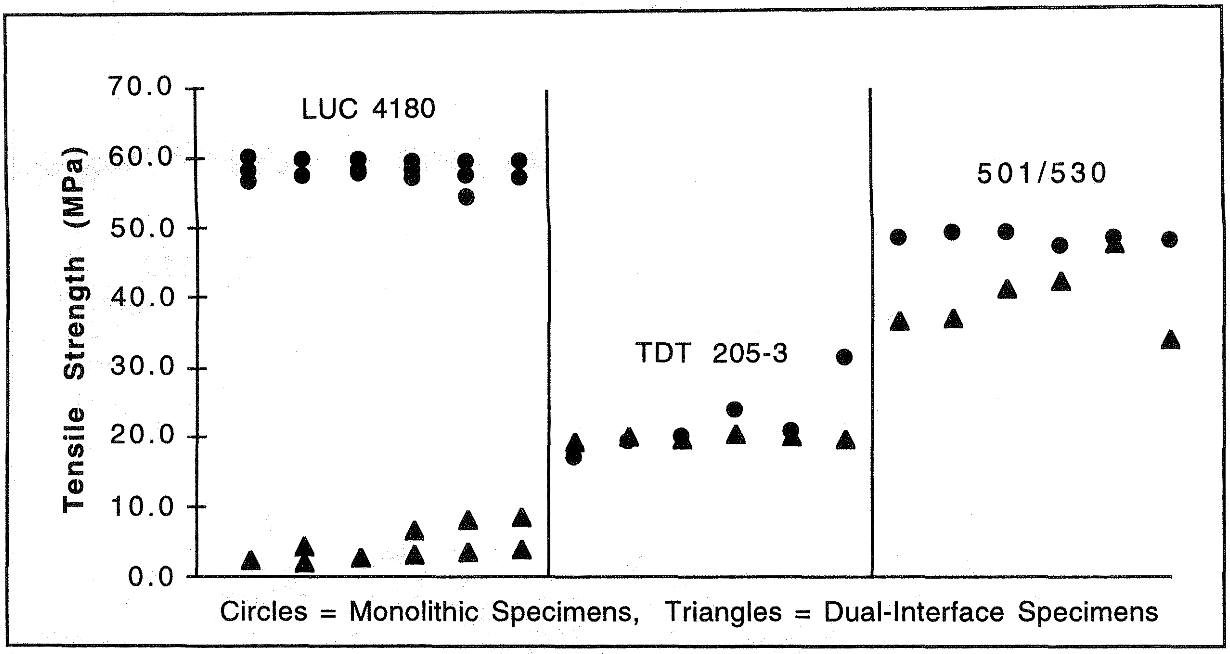

Figure 6. Tensile Strength Summary

Discussion

While monolithic LUC 4180 polyurethane is reasonably strong, dual-interface specimens of this material failed at very low stresses during tensile testing. Some layered LUC 4180 parts have suffered from delamination during handling, indicating that this problem also arises during actual use of the material. These results indicate that LUC 4180 is only useful for monolithic parts, such as those produced in the Mold SDM process [4].

Dual-interface specimens of both TDT 205-3 polyurethane and 501/530 epoxy failed at much higher tensile stresses - these two materials are better suited for producing layered parts. While TDT 205-3 has a lower interlayer strength than 501/530, it does have some advantages over that material for use in the SDM process. It has very low viscosity and good air-release properties, both of which help prevent voids in interior corners of layers. More importantly, it can be machined almost ten times sooner than 501/530, in two hours rather than twenty-four. For these reasons, TDT 205-3 is currently the polymer material of choice except for situations where the strongest possible parts are needed.

These test results demonstrate that there can be a very substantial anisotropic variation of mechanical properties in polymer Shape Deposition Manufacturing. "Relative interface strength" can be defined as the ratio of interlayer tensile strength to intralayer tensile strength; the relative interface strengths of the materials tested are reported in Table 3. Relative interface strength is a measure of strength anisotropy since it should be proportional to the difference in strengths between tensile tests parallel to and perpendicular to the build direction. For LUC 4180, tensile strength in the build direction may be twelve times lower than tensile strength within a single layer.

Both bulk strength and relative interface strength must be considered when selecting polymer part materials for SDM. However, neither a relative interface strength nor an absolute interlayer strength is normally tabulated by manufacturers since few applications require materials to adhere well when cast over machined surfaces of the same material. Maximizing interlayer strength results in the strongest objects although it can produce parts with greater anisotropy. An example is that 501/530 forms stronger interlayer interfaces than TDT 205-3, even though its tensile strength is more anisotropic.

	LUC 4180	TDT 205-3	501/530
Relative Interface Strength	8 %	90 %	82 %

Table 3. Relative Interface Strengths

As a comparison between LUC 4180 and TDT 205-3 indicates, relative interface strengths can vary significantly even within a class of materials. Both of these materials are two-part, castable, MDI-based polyurethane resins, but their relative interlayer strengths are 8% and 90%, respectively. The reasons for such differences are not well understood. The Adsorption model of adhesion is the most generally accepted theory of adhesion [10]; it suggests that complete wetting of a surface is required to form strong adhesive bonds [11]. Wetting of crosslinked surfaces by chemically similar liquids is a complex phenomena, and the degree of wetting can be affected by the degree of crosslinking of the solid surfaces [12]. Both LUC 4180 and TDT 205-3 have complex systems of chemical additives, including plasticizers, antifoam agents, and particulate fillers. All three of these components affect the wetting behavior of the materials: plasticizers directly improve wetting [13], antifoam agents reduce surface tension, and fillers raise viscosity (which can limit wetting [14]). Differences in the degree of crosslinking of the cured surfaces or wetting characteristics of the uncured liquid resins are thought to determine the relative interface strengths of these two materials by affecting their wetting behavior.

Future work will focus on the identification of stronger materials with good interlayer bonding for the production of functional parts via polymer Shape Deposition Manufacturing.

Acknowledgments

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References:

[1] R. Merz, F. B. Prinz, K. Ramaswami, M. Terk, and L. E. Weiss, "Shape Deposition Manufacturing," *Proceedings of the 1994 Solid Freeform Fabrication Symposium,* The University of Texas at Austin, August 1994, pp. 1-8.

[2] F. Prinz and L. Weiss, "Method for Fabrication of Three-Dimensional Articles," U. S. Pat. No. 5,301,415, April 12, 1994.

[3] J. W. Kietzman, A. G. Cooper, L. E. Weiss, L. Schultz, J. L. Lombardi, and F. B. Prinz, "Layered Manufacturing Material Issues for SDM of Polymers and Ceramics", *Proceedings of the 1997 Solid Freeform Fabrication Symposium,* The University of Texas at Austin, Austin, Texas, August 1997, pp. 133-140.

[4] A. G. Cooper, S. K. Kang, J. W. Kietzman, F. B. Prinz, J. L. Lombardi, and L. E. Weiss, "Automated Fabrication of Complex Molded Parts Using Mold SDM," *Proceedings of the 1998 Solid Freeform Fabrication Symposium*, The University of Texas at Austin, Austin, Texas, August 1998.

[5] L. Weiss, F. Prinz, G. Neplotnik, K. Padmanabhan, L. Schultz, and R. Merz, "Shape Deposition Manufacturing of Wearable Computers," *Proceedings of the 1996 Solid Freeform Fabrication Symposium*, The University of Texas at Austin, Austin, Texas, August 1996, pp. 31-38.

[6] "LUC-4180 Lowex Casting System," Adtech Plastic Systems Corp., Charlotte, Michigan.

[7] "TDT 205-3 Resin and Hardener / DT 081 / DT 082 Three Component Rapid-Curing Polyurethane Casting System," Ciba Specialty Chemicals Corporation, East Lansing, Michigan, October 1996.

[8] "System^5 Epoxy Encapsulation Systems," Adtech Plastic Systems Corp., Charlotte, Michigan.

[9] "Standard Test Method for Tensile Properties of Plastics," ASTM D 638-96, *1997 Annual Book of ASTM Standards*, Vol. 08.01, West Conshohocken, Pennsylvania: ASTM, 1997, pp. 44-56.

[10] A. J. Kinloch, "Review: The Science of Adhesion - Part 1. Surface and Interfacial Aspects," *Journal of Material Science*, Vol. 15, 1980, pp. 2141-2166. Page 2156 describes the adsorption theory.

[11] Georges Fourche, "An Overview of the Basic Aspects of Polymer Adhesion. Part I: Fundamentals," *Polymer Engineering and Science*, Vol. 35, No. 12, June 1995, pp. 957-967. Pages 958ff describe the adsorption theory.

[12] T. Kerle, R. Yerushalmi-Rozen, and J. Klein, "Cross-Link-Induced Autophobicity in Polymer Melts: A Re-Entrant Wetting Transition," *Europhysics Letters*, Vol. 38, No. 3, 1997, pp. 207-212. This article describes the transitions from complete wetting, to partial wetting, and back to complete wetting as a surface's degree of crosslinking was increased from none, to partial, to complete.

[13] J. K. Sears and N. W. Touchette, "Plasticizers," in eds. Herman F. Mark, Norbert M. Bikales, Charles G. Overberger, and Georg Menges, *Encyclopedia of Polymer Science and Engineering*, Supplement Volume, New York: Wiley-Interscience, 1985, pp. 568-647. Page 614 describes the effects of plasticizers in thermoset materials.

[14] A. J. Kinloch, "Review: The Science of Adhesion - Part 1. Surface and Interfacial Aspects," *Journal of Material Science*, Vol. 15, 1980, pp. 2141-2166. Page 2142 describes the qualities of an ideal adhesive, specifying a viscosity of a few centipoise.