

FABRICATING ADHESIVELESS POLYIMIDE MULTILAYER FLEX CIRCUITS

**Nancy Holloway
Electronics Engineering Technician
NASA Langley Research Center
MS 390, Hampton, VA 23681-1001**

INTRODUCTION

Future electronic systems for aeronautical, space, and consumer applications will have to be smaller, lighter, require more efficient circuitry and survive harsher environments. As these electronic systems become increasingly more complex, the requirements are driven towards denser circuitry and higher layer count (multilayer) circuits. To meet these needs, designers will have to increasingly rely on lightweight multilayer flexible circuits and cables made from higher performance materials.

Flex circuits have been produced on numerous materials, from aramid paper to very thin flexible fiberglass. Currently, most flex circuits are produced on either polyester or polyimide film. Polyester, being economically feasible to produce and having favorable dielectric properties, makes it a widely used material in the flex circuit industry. The main disadvantage to using polyester is its inability to withstand high temperatures; therefore making soldering a difficult process.¹ Polyimide, a premium thermoplastic, can be exposed to elevated temperatures, numerous chemicals, and be repeatedly soldered without degradation.² The main disadvantage to using polyimide film is the greater cost compared to polyester. Polyimide film, because of its high performance characteristics, is almost always used to produce military and aerospace flex circuits.

CURRENT METHOD OF FABRICATING MULTILAYER FLEX CIRCUITS

While there are many variations on how to fabricate multilayer flex circuits, conventional art teaches the following process.³⁻⁵ Copper clad films, which make up the multilayer flex circuit, are drilled with tooling holes. All of the films which make up the circuit are patterned, except the outermost films which are patterned after plating has taken place. Cover layers are laminated onto all of the patterned films. The films, both patterned and the outermost unpatterned, are stacked and adhesively bonded together with flexible bond ply using a laminating press. Next, the laminate is drilled and the holes are cleaned to remove drill smear and contaminates. The holes are then plated and the outermost films are patterned. Lastly, a cover layer is laminated to the outermost patterned films creating the fabricated flexible multilayer circuit board or cable.

ADHESIVES ROLE IN THE MANUFACTURE OF FLEX CIRCUITS

Most polyimide materials used to fabricate multilayer flex circuits are available in sheet form which necessitates the use of a flexible bond ply (adhesive) to join the patterned films together to create multilayers. Adhesives play an active role in fabricating multilayer circuits and are used to bond the conductor and cover layers to the film as well as bond the patterned films together to form a multilayer circuit. Several adhesives which are currently being used for flex circuits are based on: polyester, acrylic, modified epoxies, phenolic, polyimide, and fluorocarbon. Adhesives are chosen to meet the necessary requirements of the flex circuit which include compatibility with the substrate and conductor layer, and survivability from the rigors of processing. While adhesives are used to fabricate multilayer flex circuits, they often create disadvantages among which are: additional materials and processing cost, thermal mismatch between materials, greater Z axis expansion, and a less flexible higher end weight product. Adhesives can promote wrinkling in the film causing voids to form, and exposure to elevated temperatures promotes adhesive failure. Additionally, adhesives place limits on the chemical and environmental exposure of the flex circuit or cable.⁶

Industry is aware of the problems adhesives can cause with flex circuits and is producing what is called, "adhesiveless" products in which no adhesive is used to bond the conductor (usually copper) to the substrate. Two elements which cultivate the use of adhesiveless substrates are the recent mil-spec change which allows flex circuits to be produced without adhesives and an increasing need for flex circuits to be smaller, thinner, and provide an overall lighter printed circuit.⁷ Large development programs to produce adhesiveless products are underway in Europe, Japan, and the United States.⁸ However most materials, including copper, have difficulty adhering to polyimide film. Companies still use adhesives when making flex circuits, even though adhesives contribute to wrinkling and degradation of the circuit. Although adhesives are being phased out for use in bonding the conductor (copper) to the substrate, adhesives are still being used to form multilayer circuits and bond cover layers. Often, polyimide adhesives are used for making multilayer and rigid flex circuits, but polyimides as adhesives tend to exhibit a low bond strength when adhered to polyimide film. Considerable research and development, primarily overseas, is going into adhesives which would effectively bond polyimides together.⁹

Hence, a self-bonding high performance polyimide which has the required dielectric properties and chemical stability could replace both the materials and processes currently in use for the fabrication of flexible electronic hardware. Recently, the scientists and engineers at NASA Langley Research Center have been developing materials and processes to fill this need. LaRC™-SI (Langley Research Center-Soluble Imide) is such a material that meets the future requirements of the flex electronics industry. This paper outlines the fabrication techniques, material properties and electronic hardware made using LaRC™-SI.

EXPERIMENTAL

The LaRC™-SI (2% offset) (Imitec, Inc.), N-methylpyrrolidione (NMP, Fisher Scientific), Toluene (Fisher Scientific), 1818 Microposit® Photo Resist (Shipley) and Microposit® Developer (Shipley) were used as received.

Procedure for Making Adhesiveless, Self-Bonding, Multilayer Flex Circuits

a. Metallization of Polyimide Film. The polyimide film was secured to a backing plate and placed in an evaporator which was evacuated to 2×10^{-5} torr. While under a vacuum, a tie layer of 200Å of nickel was evaporated to the film, then 2,500 Å of copper was evaporated on top of the nickel to produce a metallized polyimide film.

b. Patterning Of Polyimide Film. Photo resist was spin coated on the metallized polyimide film at 1,000 rpm for 30 seconds and baked at 90°C for 30 minutes in an oven. The coated, metallized polyimide film was removed from the backing plate. A black on clear polyester sheet containing the artwork pattern (circuitry) was vacuum held against the resist coated, metallized polyimide film and exposed for 35 light units under an ultraviolet light. The film was placed in a dish filled with a solution of 100 mL of developer and 100 mL of water. The film was agitated for 1 minute yielding a patterned film. The patterned film was removed from the dish and rinsed under running water for 3 minutes. Excess nickel and copper were etched from the patterned film by pouring a solution of 400 mL water, 200 mL sulfuric acid and 10 mL hydrogen peroxide, heated to 71°C over the patterned film. The patterned film was rinsed under running water for 3 minutes. The residual photo resist was removed from the patterned film by soaking the film in acetone, then rinsing it under running water for 3 minutes. The wet patterned film was blotted dry with a lab tissue.

c. Drying of Patterned Polyimide Film. The patterned film was secured to a backing plate and dried at 100°C for 1 hour in an oven.

d. Preparation of LaRC™-SI Spray Solution. In a 100 mL Pyrex beaker, 4 grams of LaRC™-SI were mixed into 60 grams of NMP and 3 grams of toluene. The mixture was stirred for 1 minute, capped and stirred for an additional 10 minutes to afford a clear brown solution. The LaRC™-SI solution was poured into a spray jar and affixed to an airbrush.

e. Masking, Spraying and Drying of LaRC™-SI Dielectric Film. All feedthrough areas and alignment targets on the patterned film were masked (covered) with an electrical plating tape. A coat of LaRC™-SI solution was sprayed on the film covering the entire surface. After spraying, the film was immediately placed in an oven and soft baked at 50°C for 15 minutes. This spraying and drying process was repeated several times. The masked areas were removed, while the areas covering the alignment targets remained on the film. The patterned, LaRC™-SI coated film was then dried at 100°C for 1 hour in an oven.

- f. Metallization and Patterning Additional Layers of Polyimide Film. Steps a through d can be repeated as necessary to build an adhesiveless multilayer flexible film.
- h. Surface Mount Part Population. Surface mount parts were bonded to the adhesiveless multilayer film with silver epoxy to yield an adhesiveless multilayer flex circuit.
- g. Cover Coat. LaRC™-SI solution was prepared as in part d. A light coat of LaRC™-SI was sprayed onto the populated flex circuit then soft baked at 50°C for 15 minutes. This process was repeated several times. After the final coat, the populated flex circuit was hard baked at 100°C for 1 hour.

RESULTS AND DISCUSSION

Several single, double and multilayer self-bonding, adhesiveless flex circuits have been fabricated using LaRC™-SI in solution form.¹⁰⁻¹³ LaRC™-SI can easily be dissolved, sprayed, and dried at relatively low temperatures (100°C), without distorting or dissolving the previous film layer. If the spray coating dissolved or distorted the film layer, it would greatly degrade both the film and circuit pattern, rendering the package useless. LaRC™-SI is a fully cured polyimide material and can be used in a powder, gel, solution, or film form. It should also be noted that because LaRC™-SI is already cured it only needs to be dried, unlike polyamic acids which must undergo complex cure cycles at much higher temperatures (250°C and higher). During cure, the polyamic acid undergoes shrinkage, generates water and chemically changes, all of which must be accounted for in processing. When spraying LARC-SI solution to a LaRC-SI film, there is no CTE mismatch between the materials because they are the same.

The process used to prepare adhesiveless multilayer flex circuits is illustrated in Figures 1-9. The first step involves securing the polyimide film to a backing plate (Figure 1). Next the film is metallized using an evaporator (Figure 2). The first layer of circuitry is patterned on the polyimide film (Figure 3). LaRC™-SI spray coating solution is prepared by mixing LaRC™-SI powder into NMP and toluene (Figure 4). Feedthroughs and targets are masked on the patterned film (Figure 5). LaRC™-SI is sprayed over the entire surface of the masked, patterned polyimide film and dried (Figure 6). Masked areas covering the feedthroughs are removed and the film is again metallized (Figure 7). The second layer of circuitry is patterned on the film (Figure 8). Surface mount parts are bonded on the film to produce an adhesiveless multilayer flex circuit (Figure 9). Additional layers can be built up by repeating the steps outlined in figures 4 through 8.

It is important to note that metals such as copper that tarnish and oxidize at higher temperatures can be used with the low temperature (50-100°C) drying LaRC™-SI solution. It was found that LaRC™-SI solution can be dried as low as 50 °C to form thin film which can then be patterned and etched with no degradation to the film¹⁴ (Figure 10). LaRC™-SI can also be applied as a wet spin coating, screened, or cast and doctored to surfaces. Once the LaRC™-SI solution is dried, it self-bonds to the layer or substrate to which it was applied, forming a monolithic structure (Figure 11).

LaRC™-SI's electrical properties are similar to that of other polyimides with a dissipation factor of 0.002 and a dielectric constant of 3.2 at 1MHz. However, LaRC™-SI's dielectric breakdown is 7230 V/mil which is higher than most polyimide materials which are about 6000 V/mil (Table 1).

LaRC™-SI has a glass transition temperature (T_g) between 248-251 °C and a Coefficient of Thermal Expansion of 46 ppm/°C from 23-150 °C and 60 ppm/°C from 150-200 °C (Table 2). These values are typical for melt processable polyimides which have not undergone any post thin film orientation.

LaRC™-SI film has excellent chemical resistance. Strips of LaRC™-SI film were wrapped around paper clips and soaked in various chemicals for 10 days at room temperature (23 °C). The films showed <0.1% weight change and were creasible when soaked in water, jet fuel, toluene, MEK, hydraulic fluid, THF, ethylene glycol, and HCl. When the film was soaked in methylene chloride, it swelled slightly and became opaque and wrinkled, but still remained tough and creasible with a weight change of <0.1% (Table 3). Most polyimides are slightly hygroscopic and absorb moisture which can adversely effect adhesion and decrease the materials dielectric properties. Polyimide films can absorb up to 3% water by weight, however LaRC™-SI films (2% offset) have <0.1% weight change, and melt extruded LaRC™-SI films (4% offset) have a 1.66% weight change (Table 4).

Biological tests performed thus far have shown no fungus growth on LaRC™-SI film (Table 5). LaRC™-SI film has an overall low permeability of oxygen and is more selective to oxygen versus nitrogen (Table 6). LaRC™-SI, having excellent solvent and biological resistance, along with low gas permeability, makes it a choice material as an environmental barrier.

While LaRC™-SI bonds to itself, it also bonds directly to various surfaces (aluminum, ceramic, glass, copper, and polyimide film) which allows for electronic circuits to be placed on/in unique parts of a system (Figure 12). Flex circuits have been produced on aluminum and copper foil by both spraying and casting the LaRC™-SI solution on the foils. Patterned LaRC™-SI films have been hot pressed to titanium (Ti, 6A1-4V), untreated aluminum foil and Kapton® polyimide film, and were found to have excellent adhesion (See Figure 13). Ceramic substrates have also been sprayed with the LaRC™-SI coating, dried, metallized and patterned. These substrates were plunged into liquid nitrogen and warmed to room temperature without any delamination of the metal pattern or LaRC™-SI coating on the ceramic substrates.

OTHER APPLICATIONS

It was found that LaRC™-SI solution could be used as an adhesive for particle spray coating applications. Various lightweight powdered materials were suspended in LaRC™-SI solution affording a powdered spray coating. Additionally, it was also found that

heavier weight powdered materials, which would not remain suspended in the LaRC™-SI solution, could still be applied to surfaces using a slightly different technique. This technique involves spraying a coat of LaRC™-SI on the surface and then spraying the powdered material on top of the LaRC™-SI. Some of the powdered materials which have been sprayed with LaRC™-SI include: bronze, ceramic, graphite, tungsten carbide, and diamond dust. These spray coatings could be used for numerous applications including: thick film, EMI shielding, thermal management on electronic components, and wear resistant coatings on parts and tools. Other current and future electronics applications incorporating LaRC™-SI include: coatings, wire insulation, cover layers, and thin film high frequency capacitors.

CONCLUSION

Many advantages can be gained from fabricating flex circuits and cables without adhesives. However, to achieve this goal, extensive materials research must be done to provide a platform where this can take place. Thus, without the proper materials, certain advances in electronic processing technology will not occur. LaRC™-SI enables self-bonding multilayer flex circuits and cables to be made thinner with an overall lighter end weight, and without the problems of wrinkling and voids. Flex circuits and cables are no longer limited to the environmental and temperature constraints of previous circuits and cables which used adhesives. Fabricating multilayer flex circuits and cables with self-bonding film is a cost effective way to produce circuits with less processing time, capital investment, and materials costs (Figure 14). The current-state-of-the-art process of having to bond adhesives to the film, join the films in a hot press, drill holes, and clean drill smear can now be eliminated.

REFERENCES

1. J. Munson, Handbook of Flexible Circuits, Ed. K. Gilleo, p.23.
2. J. Munson, Ibid, pp. 21-23.
3. K. Gilleo, Ibid, p.11-13.
4. J. Munson, Ibid, p. 30.
5. J. F. Fjelstad, K. Gilleo, Ibid, pp. 61-62.
6. Meschter, "New Findings in Not-So-New-Wrinkling," Printed Circuit Fabrication, Vol 17, 83-86 (1994).
7. Sallo, Jerome, "Adhesiveless Flex Substrates: The whys and how," Printed Circuit Fabrication, Vol. 17, 19-20 (1994).
8. J. Munson, Handbook of Flexible Circuits, Ed. K. Gilleo, p. 33, New York, Van Nostrand Reinhold (1992).
9. J. Munson, Ibid, Ch. 2, p. 32.
10. R. G. Bryant, Patent Appl. # NASA LAR 15205-1 and LaR 15205-2, Dec. 1994.
11. R. G. Bryant, Polym. Prepr., 35(1), 517 (1994).
12. R.G. Bryant, N.M. Kruse, R.L. Fox, S.Q. Tran, Patent Appl. #NASA LAR 15387-1.

13. N.M.Kruse, A Novel Method of Fabricating Flexible Multilayer Circuits, Technology 2005, Chicago, IL, Oct. 1995.
14. S. Q. Tran, N. M. Kruse, Patent Appl. #NASA LAR 15352-1

Table 1.

**Electrical Properties of Melt
Extruded LaRC™-SI Film (4% offset)**

Measurement	Test	Result
Dielectric Breakdown	ASTM D-149	7230 V/mil
Dielectric Constant	IPC-TM-650 2.5.5.3	3.2 (1MHz)
Dissipation Factor	IPC-TM-650 2.5.5.3	0.002 (1MHz)
Volume Resistivity	IPC-TM-650 2.5.17	1.4 X 10 ¹⁶ Ω cm
Surface Resistivity	IPC-TM-650 2.5.17	6.2 X 10 ¹³ Ω
Flammability	UL 94	VTM-O rated

† Data From Trace Labs, Linthicum, MD

Table 2.

Thermal and Physical Properties of LaRC™-SI

Property	Method/Sample	Result
Inherent Viscosity	Solution>(*PAA)PI-3% offset	(0.40).33 dL/g
Physical State	Xray, DSC/Film & Powder	Amorphous
Glass Transition Temp.	DSC/Molding	248-251°C/478- 484°F
Density at 23°C	Density Column/Film	1.376 g/cm ³
Hardness (HK50)	ASTM-384-89	23-27
†Thermal Conductivity	ASTM-C-518	0.244 W/m.K
CTE (23-150°C)	TMA/Thin Film	4.6 X 10 ⁻⁵ /°C 6.0 X 10 ⁻⁵ /°C

*PAA = Polyamic acid, PI = Polyimide

†Data From Trace Labs, Linthicum, MD

Table 3.**Solvent Resistance of LaRC™-SI (2% offset)**

Solvent	Weight Change	Appearance
Water	<0.1%	NCC, Creasible
Jet Fuel (JP-5)	<0.1%	NCC, Creasible
Toluene	<0.1%	NCC, Creasible
MEK	<0.1%	NCC, Creasible
Methylene Chloride	<0.1%	NCC, Wrinkled, Creasible
Hydraulic Fluid	<0.1%	NCC, Creasible (TCP Based)
THF	<0.1%	NCC, Creasible
Ethylene Glycol	<0.1%	NCC, Creasible
HCl (Conc)	<0.1%	NCC, Creasible

Test for 10 days at room temperature on 1 mil films twisted around a tight radius.

NCC = No Color Change

Table 4.

**Unoriented Thin Film Properties
of Extruded 4% Offset LaRC™-SI**

Solvent	Weight Change*	Appearance
Water	1.66%	NCC, Creasible

*IPC-TM-650 2.6.2

Table 5.

**Fungus Resistance of a Melt Extruded
LaRC™-SI Film (4% offset)**

Fungi	Result
Aspergillus niger	No Growth
Aureobasidium pullulans	No Growth
Gliocladium virans	No Growth
Penicillium funiculosum	No Growth
Chaetomium globosum	No Growth

Data From Trace Labs, Linthicum, MD

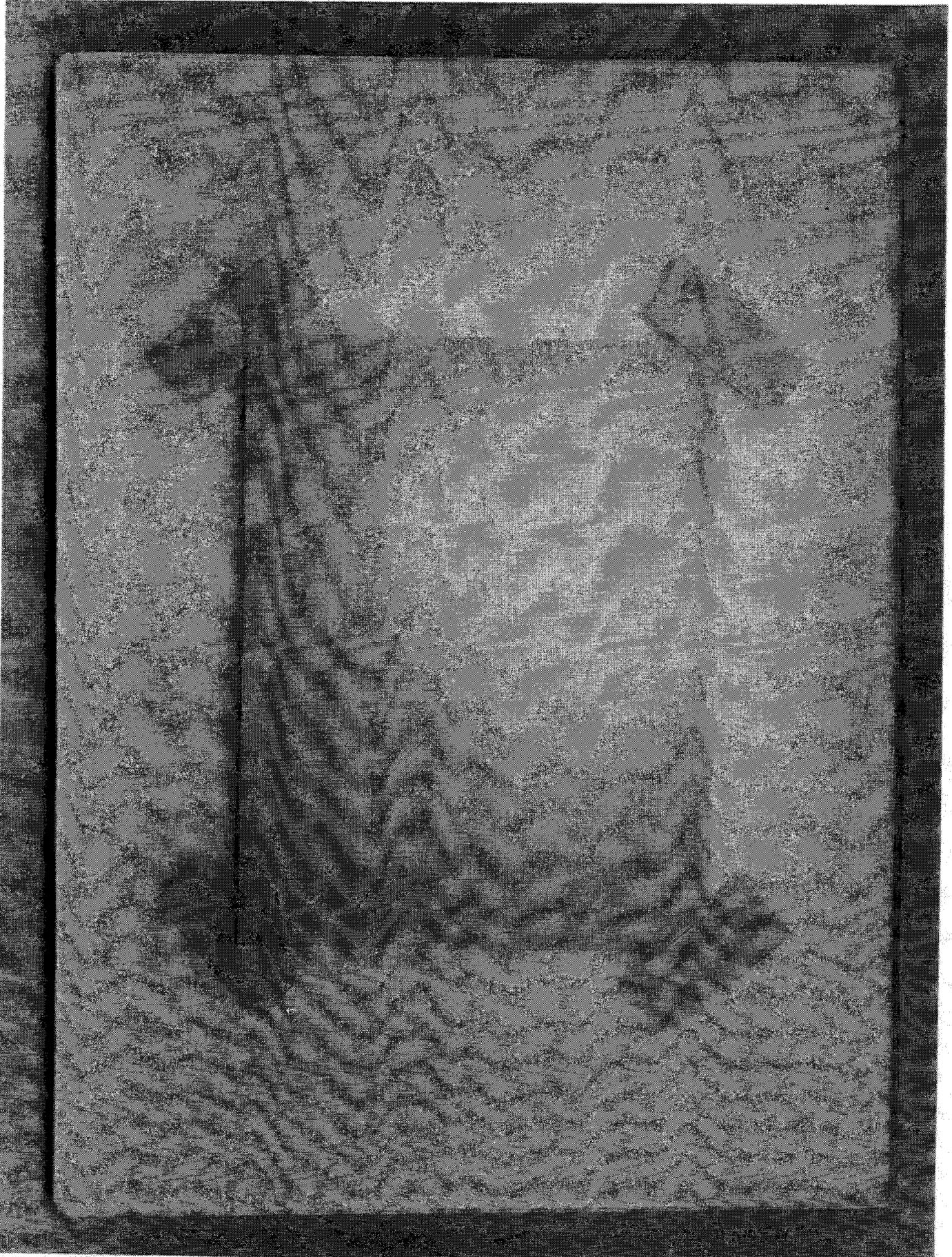
Table 6.

**Gas Permeability of LaRC™-SI (2% offset)
Unoriented Thin Films**

Method	Test Temp. °C (°F)	Result
Permeability	27 (81)	$P(O_2)=0.09$
Selectivity	27 (81)	$\alpha(O_2/N_2)=8.90$

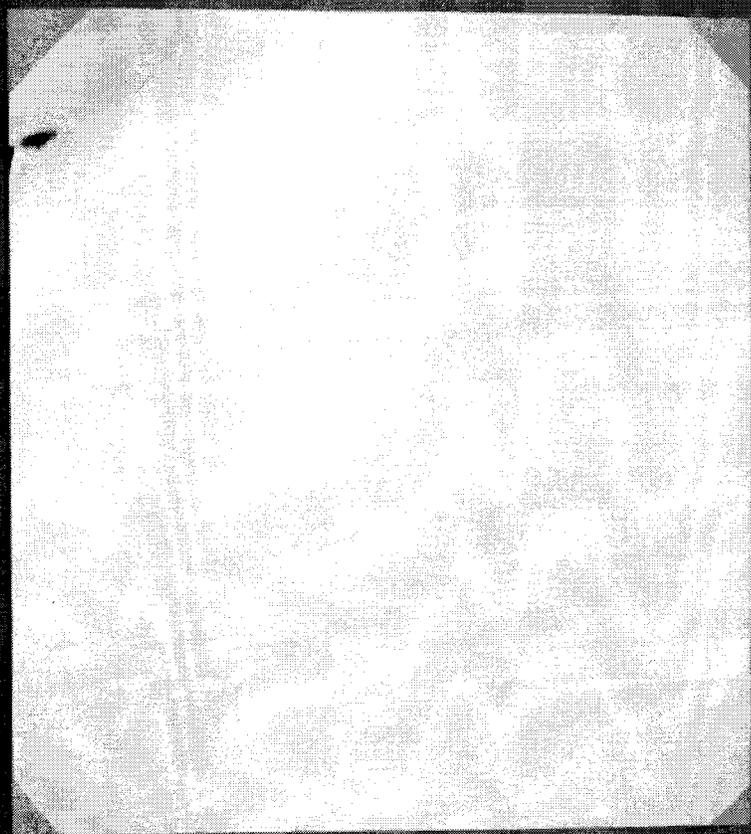
Data From AIR Products and Chemicals, Inc., Allentown, PA

Figure 1. Secure polyimide film to backing plate



96-01096

Figure 2. Metallize polyimide film



98-01091

Figure 3. Polyimide film is patterned w/first layer of circuitry

98-01042

Langley Research Center
Hampton, Virginia 22061-2100

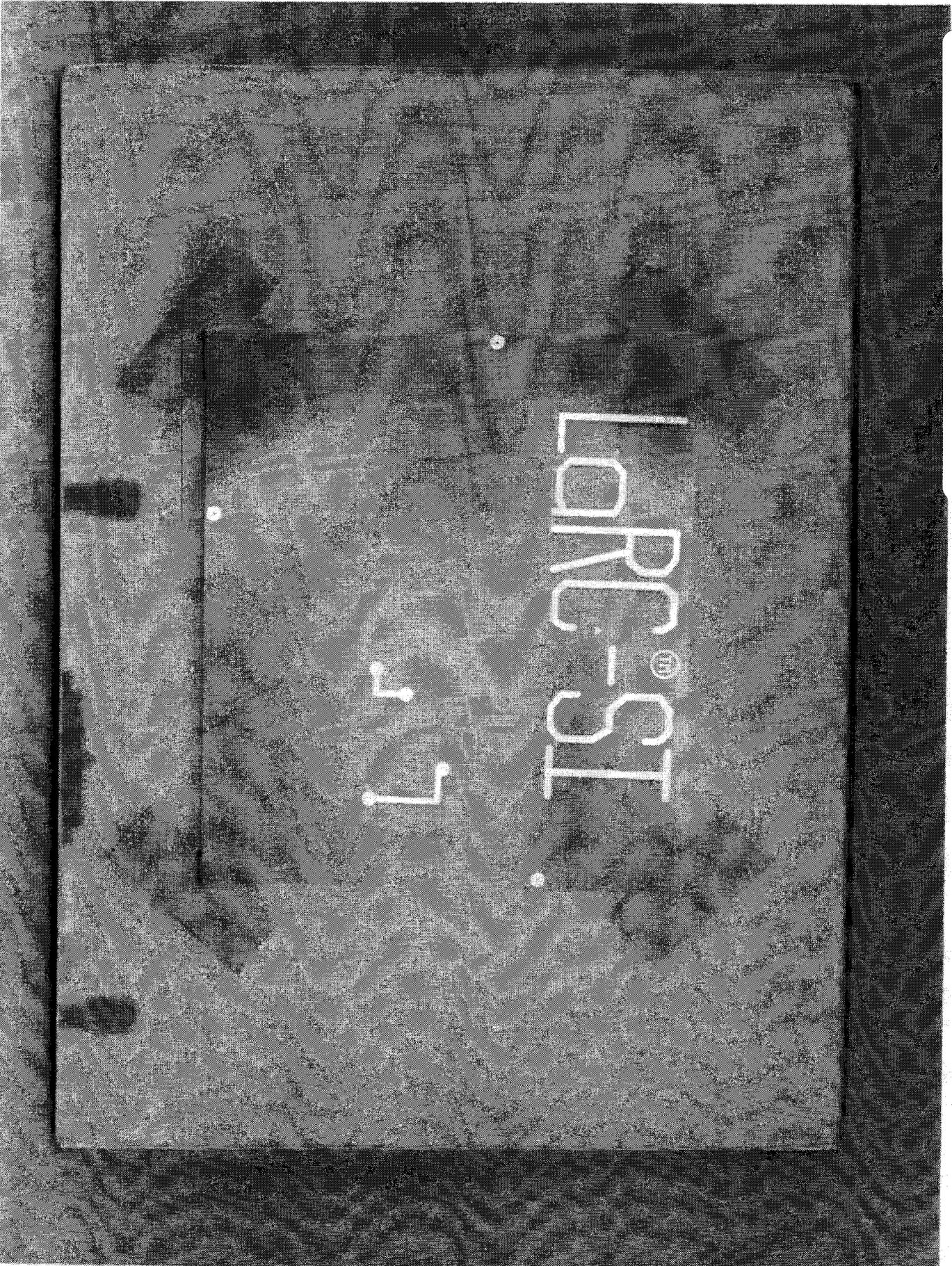


Figure 4. Prepare LaRC-SI solution
Powder mix w/NMP

95-03290

Lough Research Center
Kingston, Virginia 22085-5228

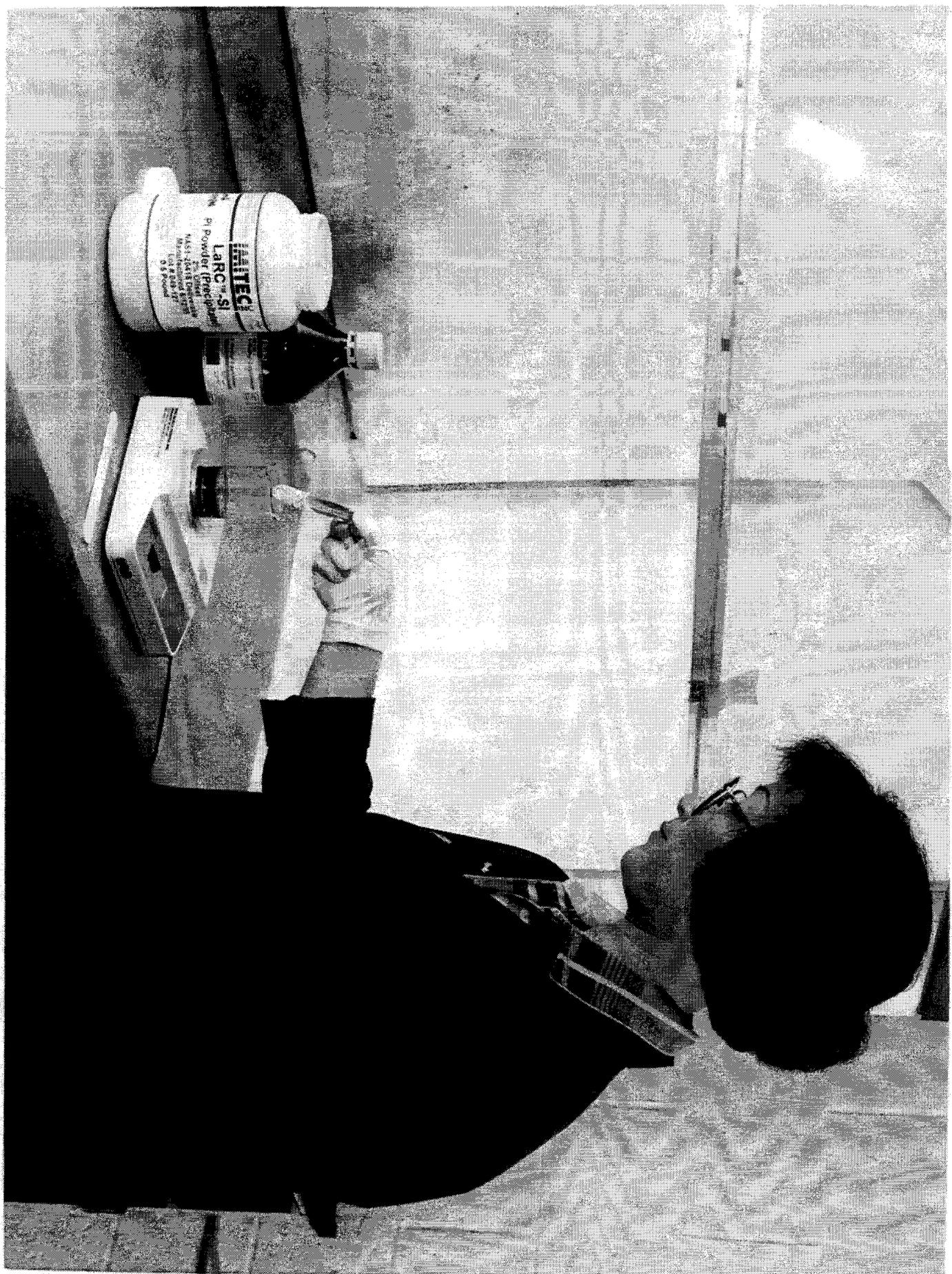
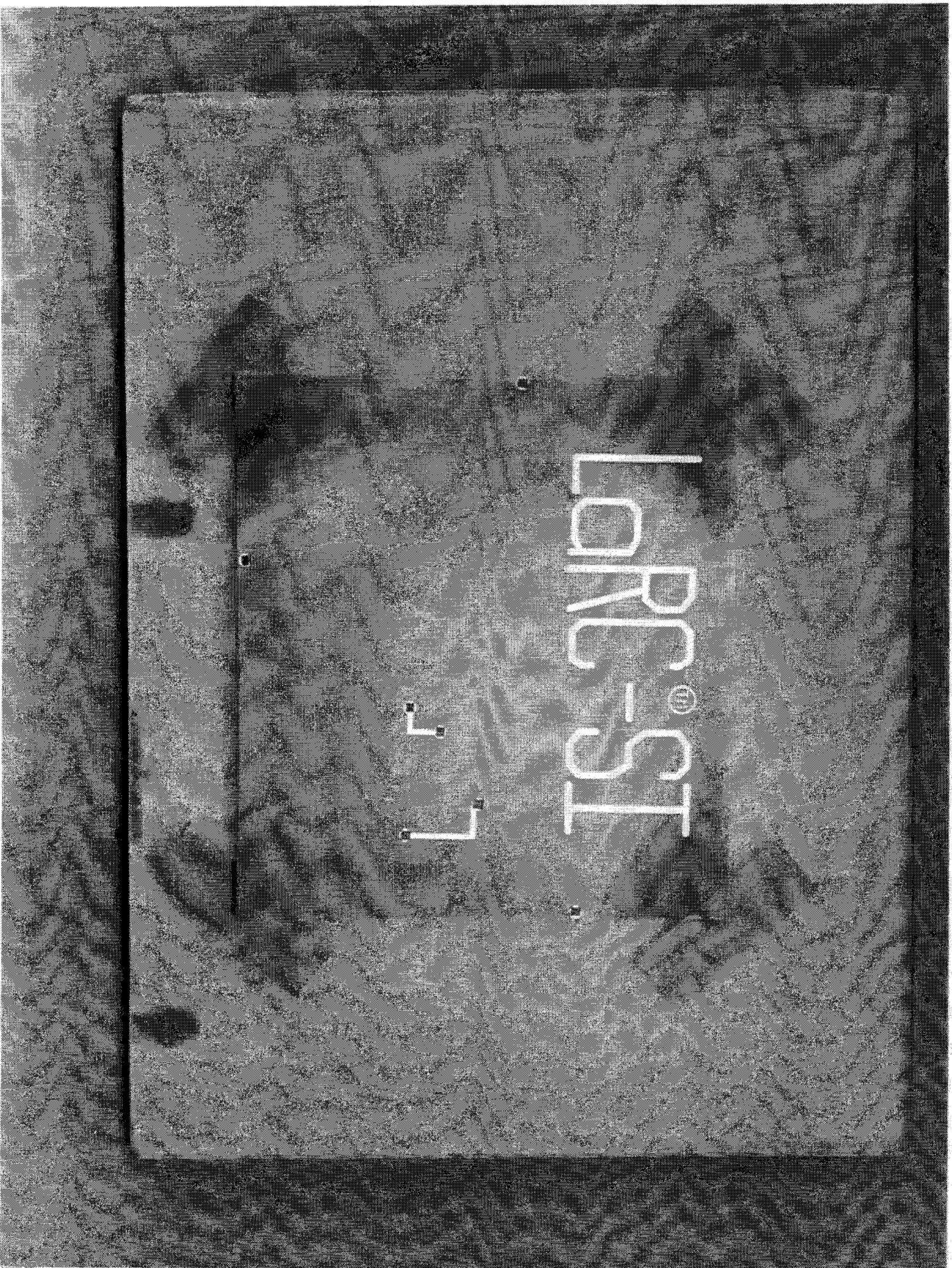


Figure 5. Feedthroughs and targets are masked



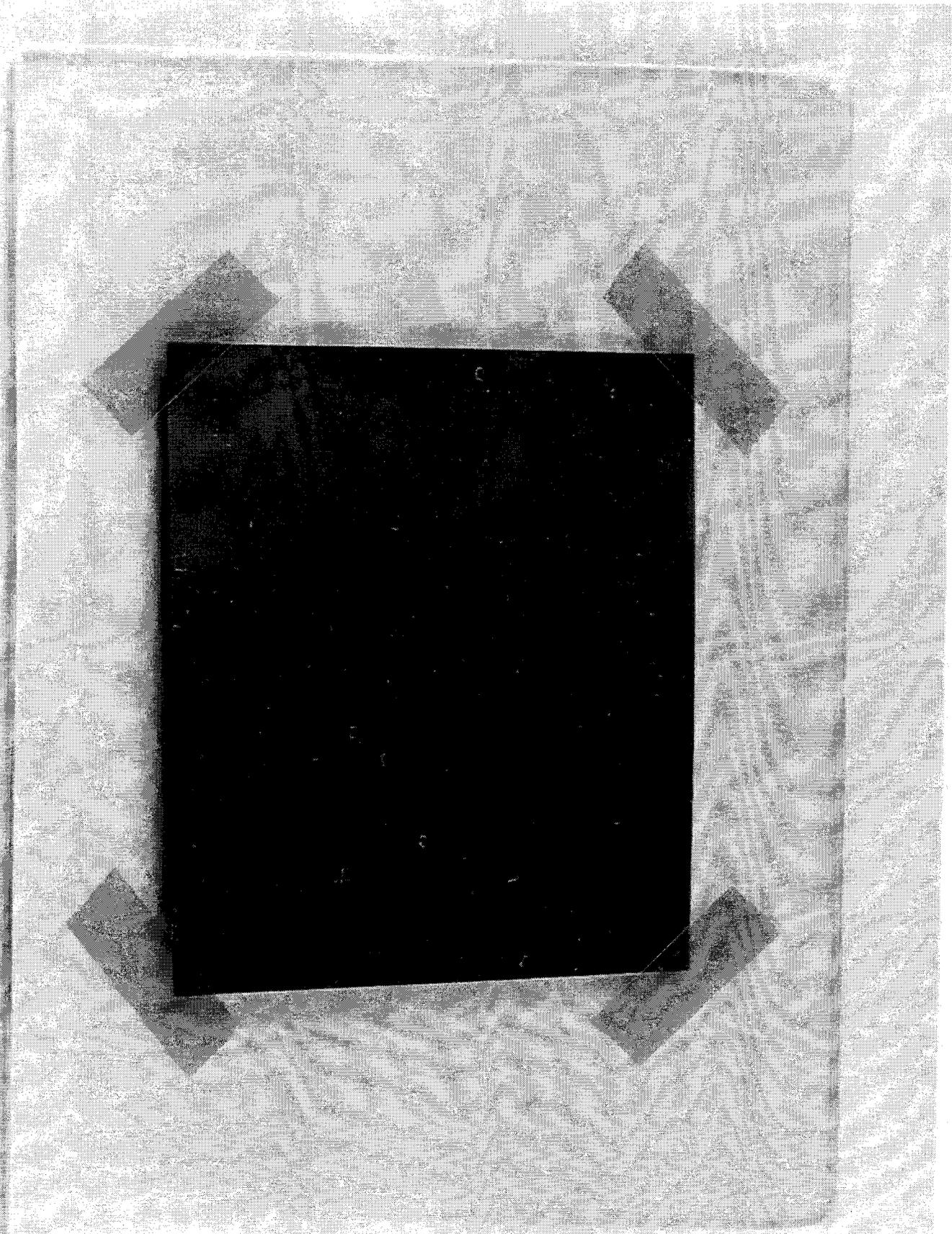
96-01043

Figure 6. LaRC-SI is sprayed over entire film surface



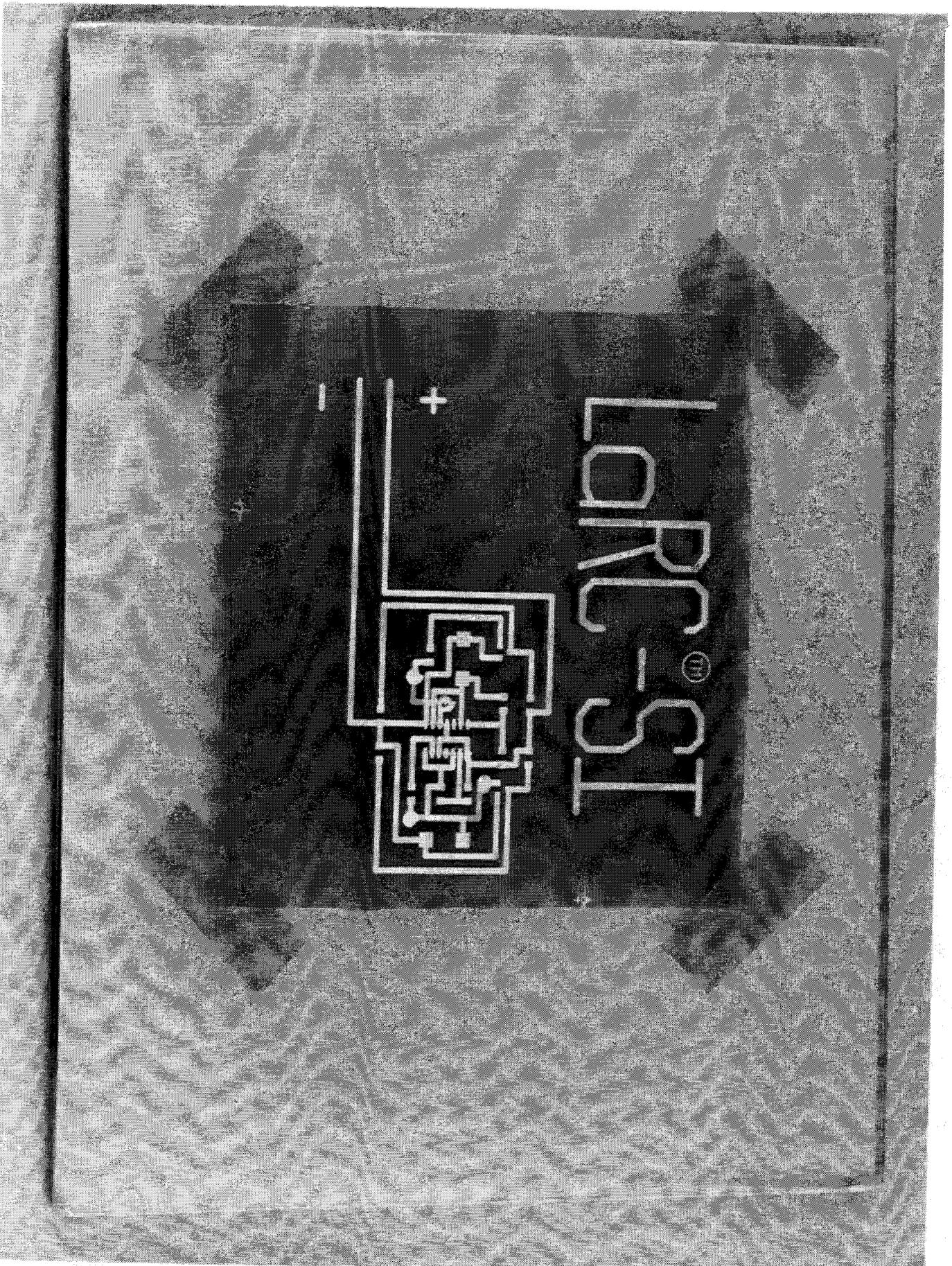
45-03000

Figure 7. Film is metallized



98-01041

Figure 8. Polyimide film is patterned w/second layer of circuitry



98-01044

Figure 9. Surface mount parts are bonded to patterned film to produce adhesiveless flex circuit

9-00915

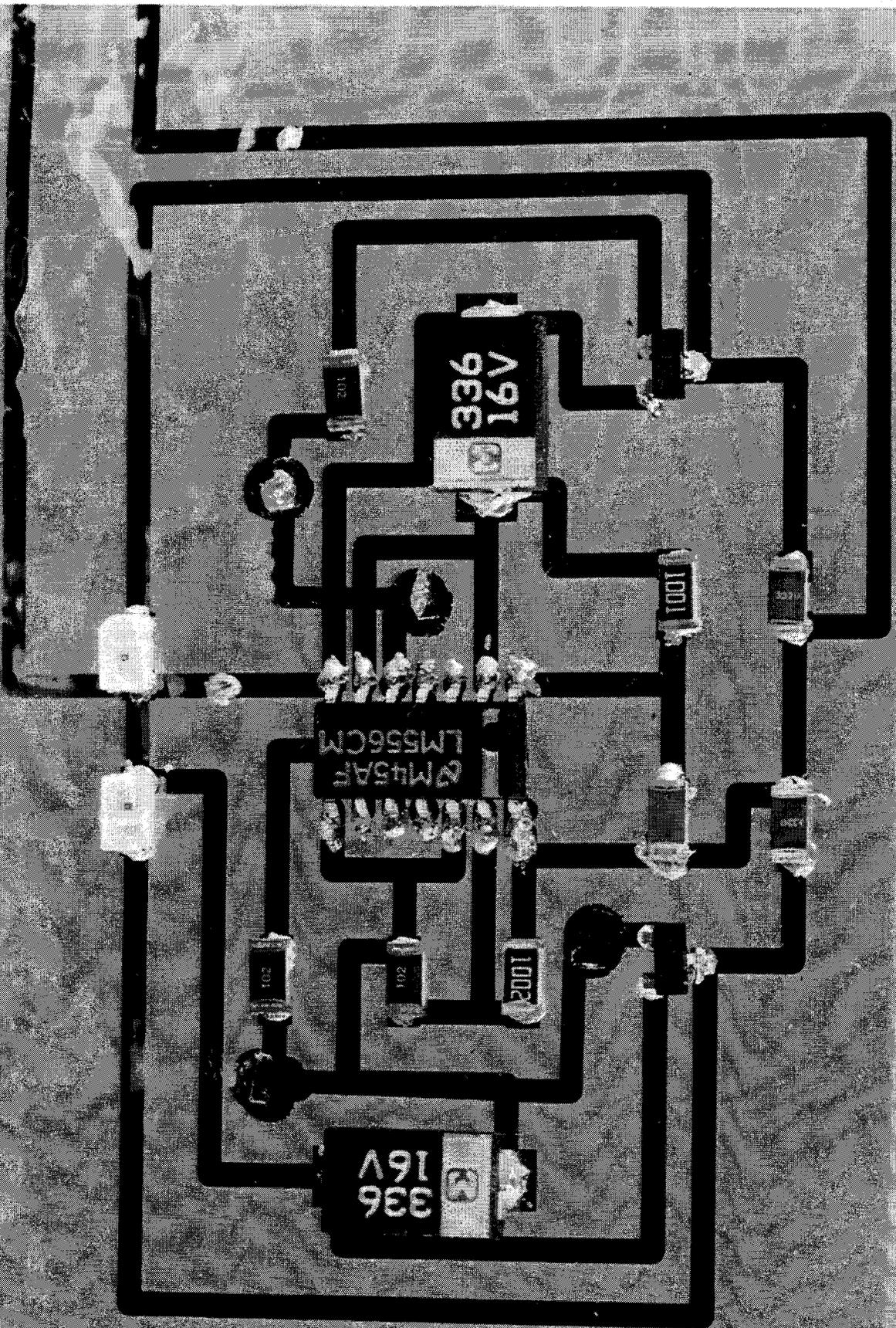


Figure 10. LARC-SI film dried at 50°C and patterned

98-00912

Langley Research Center
Hampton, Virginia 22083-2075

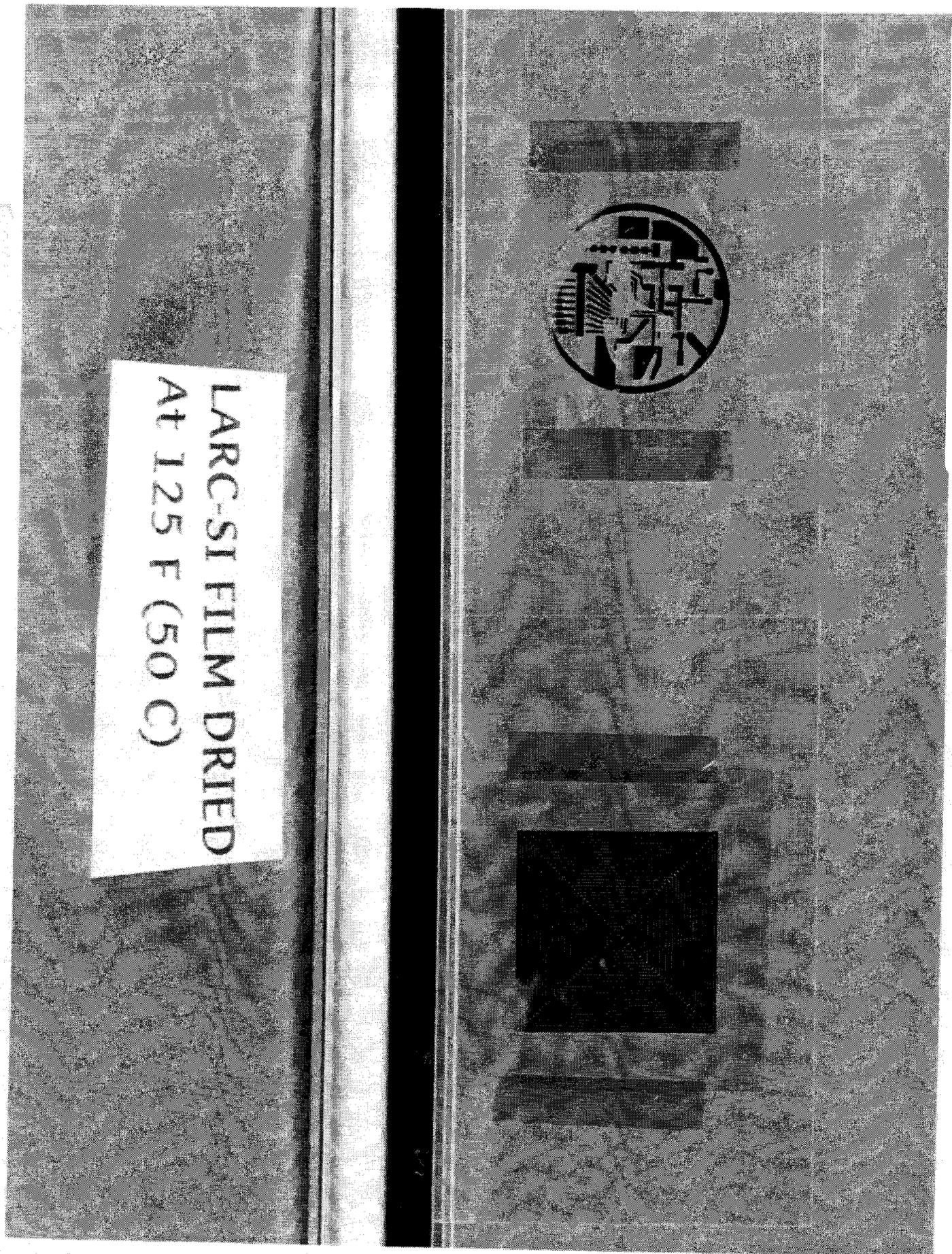


Figure 11. LaRC-SI self-bonding coating forms a monolithic structure

95-08522

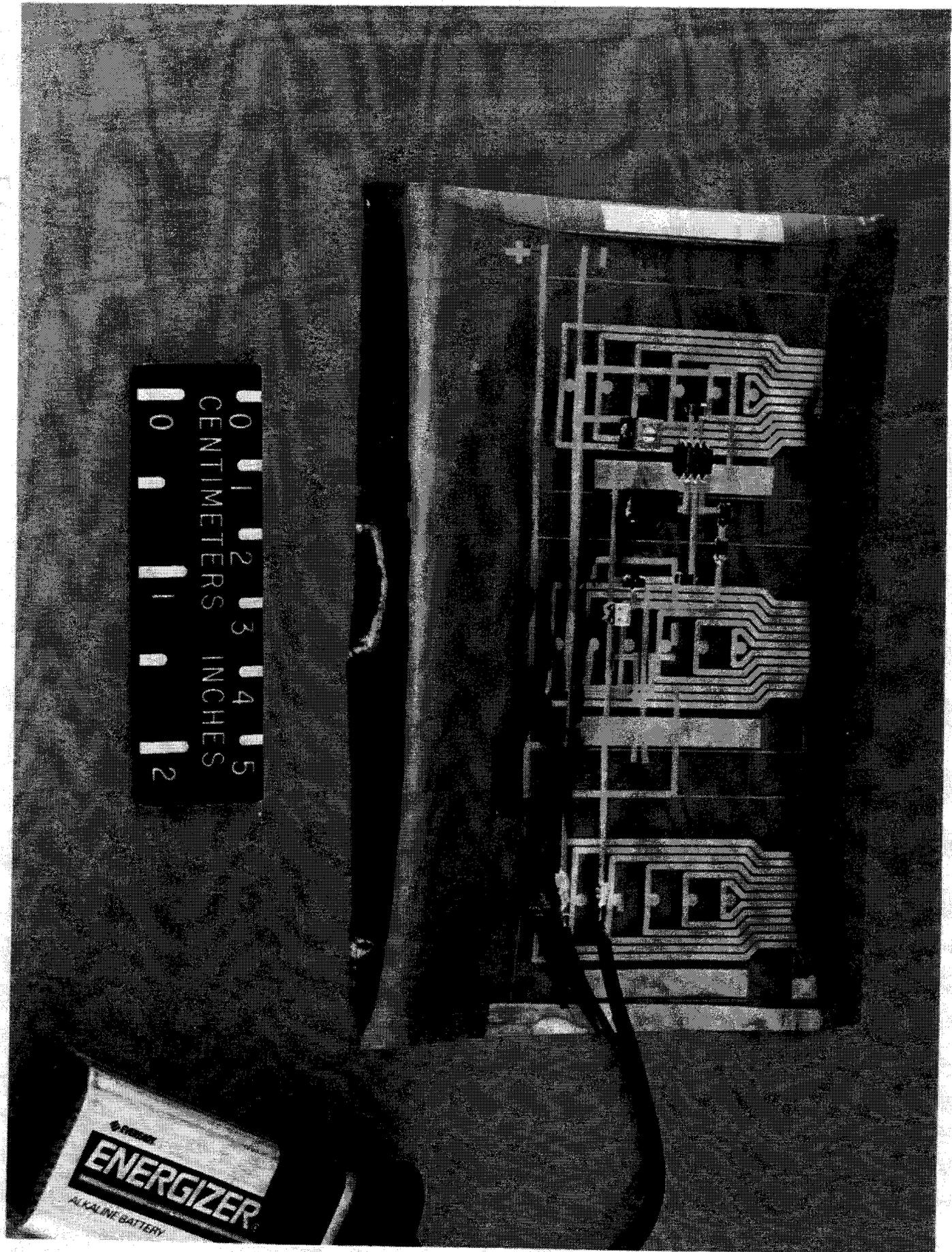
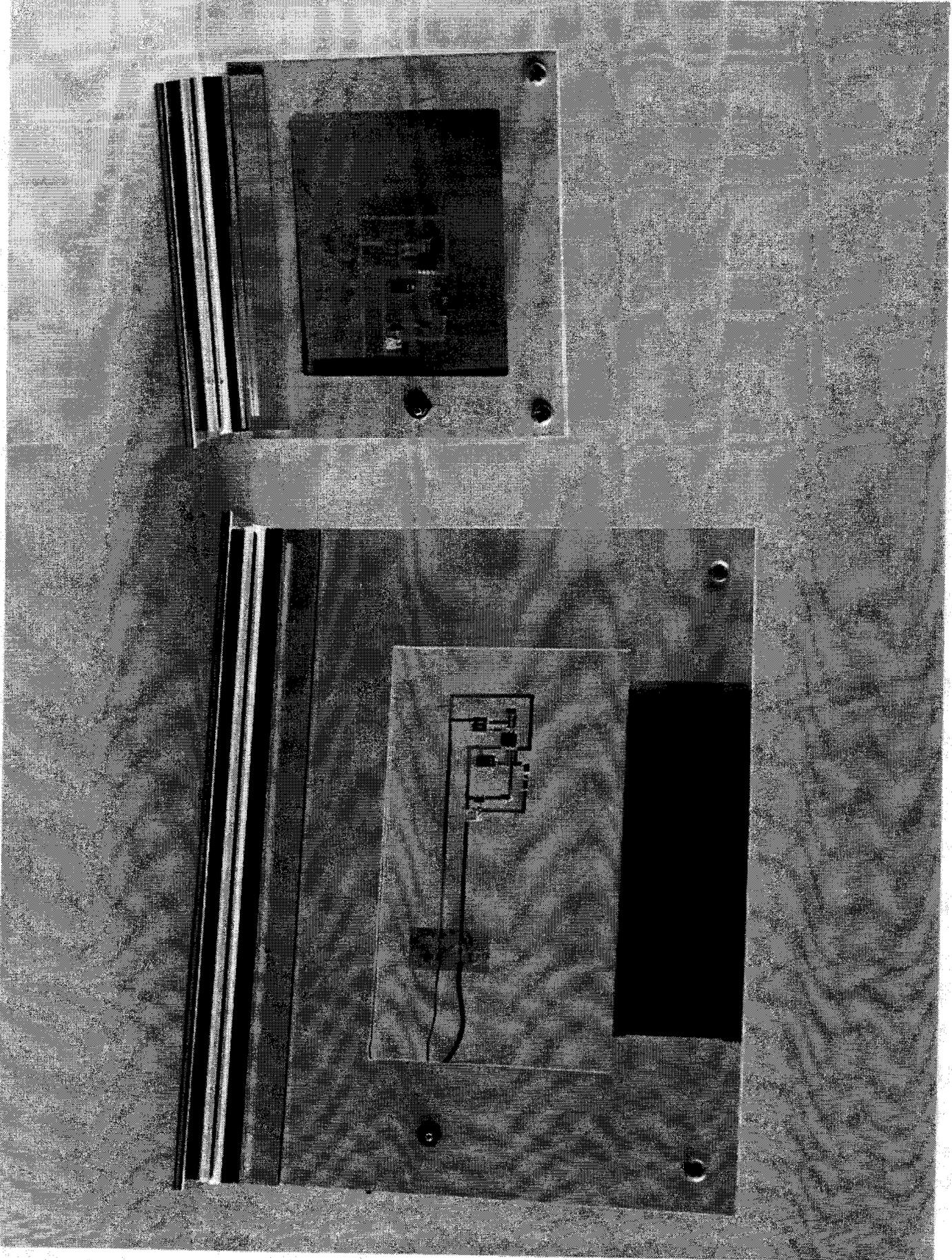


Figure 12. LaRC-SI film hot pressed to aluminum foil (right) Single sided, free standing LaRC-SI flex circuit (left)



45-00898

LA-1000-1000

Figure 13. LaRC-SI spray coatings on various surfaces: ceramic, aluminum foil, aluminum plate, LaRC-SI

45-03109

Langley Research Center
Hampton, Virginia 22083-0001



Figure 14. Adhesiveless flex circuits can be produced cost effectively utilizing IARC-SI material



94-08520

Source: Defense Science and Engineering Research Agency