CR 151224

DEVELOPMENT AND PRODUCTION OF A FLAME RETARDANT, GENERAL PURPOSE, PRESSURE SENSITIVE ADHESIVE TAPE

Final Report To

Materials Technology Branch
National Aeronautics and Space Administration
Lyndon B. Johnson Space Center
Houston, Texas 77058

By

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Arthur D. Little, Inc.

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I. SUMMARY

A. INTRODUCTION AND PURPOSE

This report summarizes the research and development work performed by Arthur D. Little, Inc. (ADL), under NASA contract NAS 9-14873. The purpose of this program was to develop and produce a nonflammable, general purpose, pressure sensitive adhesive tape.

According to the Statement of Work Paragraph 2.2, the specifications for the finished tape properties were as follows:

a. Adhesive strength (180° peel) on aluminum from 107 to 143 grams per centimeter (0.6 to 0.8 pounds per inch).

b. Adhesive strength (180° peel) on stainless steel from 71 to 107 grams per centimeter (0.4 to 0.6 pounds per inch).

c. Unwind resistance of 536 to 714 grams per centimeter (3 to 4 pounds per inch).

d. Tensile strength minimum of 7143 grams per centimeter (40 pounds per inch).

e. Elongation from 5 to 10% at break.

f. Tear strength, Elmendorf from 200 to 350 grams (0.44 to 0.77 pounds).

g. Tear strength, tongue from 363 to 408 grams (0.8 to 0.9 pounds).

h. Shelf life - no greater than a 50% increase in items a, b and c above in one year, as determined by accelerated age testing.

i. Light in color and of such a texture that it can be written upon with pencil or conventional ball-point pen.

j. Nonflammability in an atmosphere of 23.8% oxygen and 76.2% nitrogen at 1019 grams per square centimeter pressure (14.5 psia). The tape must be self-extinguishing, using bottom ignition, under the conditions specified. The flammability test shall not be LOI (limiting oxygen index).

B. SCOPE

The technical tasks performed, as specified in the Statement of Work Paragraph 2.1, were the selection and development of the components for an all-purpose utility tape consisting of:

a. A self-extinguishing substrate material;

b. A self-extinguishing adhesive to be applied to the substrate;

c. A primer, if required, with good flame resistance to secure the adhesive coating to the substrate;

d. A backsizing (release coating) with good fire resistance to be applied to the back of the tape substrate; and

e. A nonflammable core on which the finished tape could be wound.

Upon the laboratory development of a suitable pressure sensitive adhesive tape, ten 5.08 centimeter (2 inch) wide by 6.1 meter (20 foot) long rolls of the tape were produced, evaluated, and delivered to the NASA Lyndon B. Johnson Space Center.

C. RESULTS

After evaluations of numerous commercial tapes, substrate materials, adhesive systems, backcoatings and cores, the following materials were ultimately selected for the production of the finished flame retardant utility tape. Nomex paper, 0.076 millimeter (0.003 inch) thick, style 410, was used as the substrate material. A flame retardant acrylic emulsion, Rhoplex HA20, was coated on one side of the Nomex paper to provide a backcoating with suitable properties necessary for smooth unwinding of the final tape. A thin primer coat of silicone adhesive was applied to the opposite side of the Nomex paper using a diluted solution of Silgrip SR529 adhesive. Flame retardant pressure sensitive adhesive formulation, ADL No. 18887-46 (Table I) was coated over the primer coat and the adhesive tape was wound onto 1.3 millimeter (0.05 inch) wall, 76 millimeter (3 inch) diameter, hollow aluminum cores.

The finished tape, ADL No. 18887-48, was tested and compared against the contract specifications (Table II) and ten 5.08 centimeter (2 inch) wide by 6.1 meter (20 foot) long rolls of tape were shipped to NASA.

TABLE I. FLAME RETARDANT ADHESIVE NO. 18887-46

Material	Percent by Weight (Total)	Percent by Weight (Solids)
Silgrip SR574	61.4	85.6
SRC30 (catalyst)	0.6	0.6
Decabromobiphenyl oxide	4.3	9.2
Antimony trioxide	2.2	4.6
Methyl ethyl ketone	23.6	--
Toluene	7.9	--
Total	100.0	100.0

TABLE II. FLAME RETARDANT TAPE NO. 18887-48 EVALUATIONS

Tape Properties	Initial Contract Specifications	ADL No. 18887-48 Tape
Adhesive Strength on Aluminum (180° Peel)	107-143 grams/cm (0.6-0.8 lb/in)	125-161 grams/cm (0.7-0.9 lb/in)*
Adhesive Strength on Stainless Steel (180° Peel)	71-107 grams/cm (0.4-0.6 lb/in)	107-143 grams/cm (0.6-0.8 lb/in)*
Unwind Resistance	536-714 grams/cm (3-4 lb/in)	680-907 grams (1.5-2.0 lb)***
Tensile Strength	Minimum of 7143 grams/cm (40 lb/in)	7500 grams/cm (42 lb/in)**
Elongation at Break	5-10%	7-8%**
Tear Strength, Elmendorf	200-350 grams (0.44-0.77 lb)	--
Tear Strength, Tongue	363-408 grams (0.8-0.9 lb)	318-408 grams (0.7-0.9 lb) (Machine Direction)
Color	Light	Light (off-white)
Printability	Printable with Pencil or Ball-Point Pen	Printable with Pencil and Ball-Point Pen
Flammability	Self-extinguishing after bottom ignition in 23.8% oxygen, 76.2% nitrogen, 1019 grams/sq. cm (14.5 psia).	Self-extinguishing after bottom ignition in 24% oxygen, 76% nitrogen, atmospheric pressure.

* Tested at 30.5 cm/min (12 in/min) on 2.5 cm wide (1 inch wide) specimen.

** Tested at 30.5 cm/min (12 in/min) on 5.1 cm wide (2 inch wide) specimen with initial clamp separation of 15.2 centimeters (6 inches).

*** Tested at 50.8 cm/min (20 in/min) on 5.1 cm wide (2 inch wide) roll.

II. PROGRAM

A. COMMERCIAL TAPE EVALUATIONS

In conjunction with evaluating the various components for a pressure sensitive tape, we conducted an extensive search and evaluation program to identify any commercially available tapes which might meet the basic requirements. No commercial tape was identified which could meet all of the contract specifications.

B. SUBSTRATE MATERIALS

1. Requirements

In evaluating and selecting substrate materials, a balance had to be obtained among the requirements for the physical strength (tear, tensile and elongation), flame resistance, color, texture, and adhesive properties of the finished tape. All of the readily available flame resistant fabrics are usually very high in physical strength and tapes produced with these fabrics would be considerably outside the specifications. Most importantly, the tear strength of such tapes would far exceed specifications and make it extremely difficult, if not impossible, to tear the tape by hand. By contrast, cotton is one of the few fabrics capable of meeting the strength, color and texture requirements, but it is among the most difficult fabrics to render flame retardant, particularly against bottom ignition and especially when the cotton is subsequently coated. The color and texture requirements (light and printable) preclude the use of many fabrics where flame retardant treatment significantly darkens the fabric.

Finally, to achieve the optimum adhesive properties from a flame retarded adhesive, the substrate must be lightweight and sufficiently flexible to permit good contact wetting of a surface by the pressure sensitive adhesive coating. The substrate must also be smooth and uniform enough to allow for an even application of the adhesive, while being free from protrusions (such as multiple fiber ends) which might penetrate through the applied adhesive and interfere with the adhesive tack and ultimate wetting and bonding of the adhesive to a surface.

2. Findings

We obtained and evaluated a number of substrate candidates. A brief description of the significant findings is as follows:

Cotton: Cotton is well suited for this application in all areas except flammability, as cotton is readily flammable. Effective flame retardant treatments for cotton work by an intumescent (char-forming) process in the solid-phase, as opposed to the vapor-phase protection of halogenated flame retardants. Vapor-phase protection is relatively ineffective due to the high fuel content of cotton and its tendency to liberate large amounts of highly flammable vapors during pyrolysis. The intumescent mechanism generally does not, however, provide adequate flame resistant protection for cotton in oxygen-enriched atmospheres. Further, we found that the coating of a flame retarded (intumescent) cotton with a flame retardant (halogenated) adhesive material interfered with the char-forming process and most, if not all, of the cotton's protection was lost. Two experimental cottons (supplied by the U.S. Department of Agriculture) with vapor-phase (halogenated) flame retardants were also evaluated and found to be far too flammable for this application.

Vinyl: Flame retardant poly(vinyl chloride) materials were evaluated and showed promise from a flame resistance standpoint, but were totally unacceptable based on tensile strength, tear strength and elongation criteria.

Miscellaneous Synthetic Fabrics: Asbestos, fiberglass, F. R. rayon, F. R. polyesters, F. R. polyester-cotton blends, F. R. nylon and high temperature aromatic polyamide (Nomex and Kevlar from duPont) fabrics were all screened, as were several aluminized versions of these fabrics. All were found unacceptable due to their excessively high tear and tensile strengths. Tapes made with these fabrics would be extremely difficult or impossible to tear by hand.

Treated Nomex Fabrics: Several treated Nomex fabrics were evaluated. The treatment involves an oxidizing-halogenating process designed to further improve the flame resistance of Nomex fabric, but it also drastically reduces the physical strength of Nomex to a level which falls within the strength specifications for the flame retardant tape. One set of such materials, Durette, produced by Fire Safe Products, Inc., consists of Nomex fabrics all woven from staple yarns (noncontinuous filaments). The effect was to have excessive unbound fiber ends which protruded through the adhesive coating and generally destroyed the adhesive properties. Even those treated fabrics which barely fell within a useful range of strength properties were extremely dark in color.

The second source of treated Nomex was a material called "Fypro," which is no longer commercially produced. A sample of Fypro fabric was supplied to us by NASA. The material was very dark in color. Experimental tape produced with this fabric was below specifications in tear strength, and the tensile strength varied considerably from one area to another, generally falling well below the 40 pounds per inch minimum.

Nomex Paper: Nomex paper is chemically identical to Nomex fabric. However, rather than being an extremely high strength woven material, it is a moderate strength nonwoven product with the same inherent high level of flame resistance. One style of Nomex paper, type 410, is a highly calendered, dense product ideally suited for use as an adhesive tape substrate. Several thicknesses were evaluated and the 3 mil thickness was found to provide the required physical strength properties, color and texture as well as the flexibility necessary for good adhesive tape performance. This material was ultimately selected for use in producing the finished tape.

C. ADHESIVE SYSTEMS

1. Requirements

When developing a flame retardant adhesive system, as with the substrate materials, a balance must be obtained among the required properties of the adhesive. The three properties of concern are the adhesive strength, flammability, and stability. In general, the addition of flame retardant additives will detract from adhesive strength and stability.

Pressure sensitive adhesives function by virtue of their ability to flow and thereby wet surfaces. Once intimate contact has been made, the adhesive must have sufficient cohesive (internal) strength and affinity for the surface to which it is applied to maintain the bond. The addition of extender materials such as flame retardants, whether liquid or pigment-type, tends to severely impair these required properties. It is, therefore, necessary to select an adhesive which tolerates the maximum amount of flame retardant additives while exhibiting inherent flame retardant properties such that minimum addition is required. At the same time a flame retardant system which is effective at a minimum level is desirable.

The stability of a flame retardant adhesive system depends on the permanent compatibility of the components, as well as the stability of the ingredients themselves. The major areas of concern are the migration of a component (usually the flame retardants) from the system, chemical reaction (attack) of one component with another, and the chemical stability of the flame retardants themselves. These problems may be avoided by utilizing pigment-type (non-soluble) solid additives, using a minimum level of these additives, selecting chemically compatible and non-reactive ingredients, and particularly by choosing thermally and environmentally stable flame retardants.

2. Materials Selections

We undertook a survey of the adhesives industry to identify and evaluate any commercially available flame retardant adhesives which might meet the requirements of this particular application. No pre-formulated pressure sensitive adhesive which met all the qualifications was found.

We next proceeded to identify an adhesive system which, when formulated with the proper flame retardant additives, would meet the adhesive tape requirements. Since it is advantageous to use adhesives which are inherently flame retardant in order to keep the level of additives to a minimum we selected a series of silicone pressure sensitive adhesives called Silgrip, produced by General Electric, for our initial evaluations. This selection was based on the fact that a silicone adhesive possesses more inherent flame resistance than do the more conventional acrylic or rubber-resin adhesives. Most acrylic and rubber-resin adhesives will burn vigorously in air, while the Silgrip adhesives, without modification, are self-extinguishing after bottom ignition in air. The silicone adhesives can therefore be made self-extinguishing in 23.8% oxygen with fewer flame retardant additives. The Silgrip adhesives also provided a substantially higher level of adhesive performance than was required by the tape specifications. This higher baseline performance provided the necessary flexibility (overkill) in properties needed to tolerate the addition of flame retardant chemicals.

Flame retardant systems selected for evaluations in the Silgrip adhesives included hydrated alumina (suggested by the adhesive manufacturer), decabromobiphenyl oxide (representative of the more stable aromatic bromine-type flame retardants and known to be effective at low levels with silicones), and chlorinated paraffins (representative of both the aliphatic chlorine-type and liquid-type flame retardants).

3. Findings

Decabromobiphenyl oxide (DBBO), FR300BA from Dow Chemical, when used in combination with antimony trioxide at a ratio of 2 to 1 by weight, was found to be far more effective than the other approaches in increasing the flame resistance of the Silgrip adhesives. Antimony trioxide and DBBO were selected as the flame retardant system for further development efforts. Extensive laboratory compounding and testing of numerous flame retardant levels in adhesive formulations were performed to establish the relationship between DBBO/antimony trioxide level and flammability. The basic results of this work are illustrated in Table III.

Even at the relatively low levels of flame retardants, the reduction in the adhesive tack and peel strength was obvious. The initial work was performed with Silgrip SR537. We subsequently changed to Silgrip SR574 which provided higher adhesive tack and peel strength. The change to SR574, and holding the DBBO level to roughly 7 parts per 100 parts of Silgrip, produced an adhesive system capable of meeting all of the adhesive tape specifications. The finalized laboratory adhesive formulation, No. 18887-44-2, is detailed in Table IV.

TABLE III. EFFECT OF DECABROMOBIPHENYL OXIDE ON ADHESIVE FLAMMABILITY

Material	Parts by Weight					
Silgrip SR574 (65% solids solution)	100	100	100	100	100	100
SRC30 (catalyst)	1	0	1	1	1	0.7
Decabromobiphenyl oxide	0	4	6	7	8	10
Antimony trioxide	0	2	3	3.5	4	5
Flammability*	21	23	24	25	27	30

* Tested on Nomex paper, backcoated with Rhoplex HA20, and expressed as approximate maximum percent oxygen in which samples would self-extinguish after bottom ignition.

TABLE IV. FINAL EXPERIMENTAL FLAME RETARDANT ADHESIVE NO. 18887-44-2

Material	Parts by Weight
Silgrip SR574	100
SRC30	1
Decabromobiphenyl oxide	7
Antimony trioxide	3.5
Methyl ethyl ketone	50

D. RELEASE COAT MATERIALS

1. Requirements

The backsizing applied to the back of the adhesive tape substrate enables the tape to be unwound from the roll by allowing the adhesive coating to release cleanly and preferentially from the back surface of the tape rather than delaminating from the face of the tape.

Materials such as alkyds, acrylics and styrene-butadiene polymers as well as chemical complex treatments (duPont Quilon) are common backsizing ingredients for adhesive tapes. The Silgrip adhesives have a relatively low surface energy and adhere well to other low energy surfaces. Acrylics typically have a rather high surface energy and were, therefore, selected for evaluation as candidate backsizings.

2. Findings

The first release coating evaluated, duPont acrylic release emulsion No. 56106, performed extremely well as a release material. The release of the adhesive tape from this backing was smooth, uniform and of the proper value to meet the unwind specification of 536-714 grams per centimeter (3 to 4 pounds per inch) width. This acrylic material, however, is not flame resistant and increased the flammability of the overall system beyond specifications. Addition of flame retardants to the duPont product met with only marginal success.

A second acrylic emulsion, Rhoplex HA20 from Rohm and Haas, was obtained for testing as a release coating. Rhoplex HA20 is a proprietary flame retardant acrylic system. This product provided the necessary combination of good release properties as well as good flame resistance. Coating the Nomex paper with roughly 0.025 millimeters (0.001 inches) of Rhoplex HA20 increased the maximum oxygen level in which the substrate would self-extinguish after bottom ignition from approximately 22% to 24%. Rhoplex HA20 was selected for use as the release coating in the production of the finished flame retardant adhesive tape.

E. CORE MATERIALS

1. Requirements

The core, around which the flame retardant tape is to be wound, has to be nonflammable. The core also should be rigid enough to support the roll structure and preferably be light in weight.

2. Findings

One readily available core material was flame retardant rigid poly (vinyl chloride) pipe. This material met the basic requirements, but was not particularly lightweight. A second material, aluminum pipe, also met the requirements and was roughly three times lighter than the poly (vinyl chloride) pipe. Hollow aluminum pipe, with a 7.6 centimeter (3 inch) diameter and a 0.13 centimeter (0.05 inch) thick wall, was selected as the core material.

F. TAPE PRODUCTION

The flame retardant pressure sensitive utility tape was manufactured on our pilot coater utilizing the following materials and procedures:

1. Substrate

A 38.1 centimeter (15 inch) wide roll of 0.076 millimeter (0.003 inch) thick, style 410 Nomex paper was used as the substrate material.

2. Release Coat

Rhoplex HA20 acrylic emulsion was used as the release coating material. The Rhoplex was applied as a 0.038 millimeter (0.0015 inch) wet film onto the Nomex paper by reverse roll coating at 61 centimeters (two feet) per minute. The coating was dried in-line in a two-stage oven at 60°C and 80°C. The residence time at each temperature was four minutes.

3. Primer Coat

Silgrip SR529 adhesive, diluted 2 to 1 with methyl ethyl ketone, with 5 phr SRC30 catalyst was used as the primer coat. A 0.038 millimeter (0.0015 inch) wet film of primer was coated onto the Nomex paper on the opposite side from the release coat. The coating was applied by reverse roll at 91 centimeters (3 feet) per minute, dried in-line for 2.7 minutes at 90°C and cured in-line for 2.7 minutes at 160°C.

4. Adhesive Coat

Flame retardant adhesive formulation No. 18887-46 (Table V) was used as the adhesive coat.

The adhesive formulation was blended on a Cowles Dissolver immediately prior to coating to prevent the settling out of the suspended flame retardant additives. A 0.076 millimeter (0.003 inch) wet film of adhesive was applied over the primer coat by reverse roll at 61 centimeters (2 feet) per minute. The adhesive film was dried in-line for four minutes at 90°C and cured in-line for four minutes at 160°C. Two lengths, approximately 7.6 meters (25 feet) each, of the cured adhesive tape were taken up onto two 7.6 centimeter (3 inch) diameter, 0.13 centimeter (0.05 inch) wall, hollow aluminum pipes in the full 38.1 centimeter (15 inch) width. The two 38.1 centimeter (15 inch) wide "rolls" of tape were subsequently cut into 5.08 centimeter (2 inch) wide individual rolls.

TABLE V. FLAME RETARDANT ADHESIVE NO. 18887-46

Material	Percent by Weight (Total)	Percent by Weight (Solids)
Silgrip SR574	61.4	85.6
SRC30 (catalyst)	0.6	0.6
Decabromobiphenyl oxide	4.3	9.2
Antimony tiroxide	2.2	4.6
Methyl ethyl ketone	23.6	-
Toluene	7.9	-
Total	100.0	100.0

III. FINISHED TAPE EVALUATIONS

The finished flame retardant utility tape, ADL No. 18887-48, was tested and compared with the contract specifications. The results of these evaluations are detailed in Table VI.

Ten 5.1 centimeter (2 inch) wide by 6.1 meter (20 foot) long rolls of the flame retardant pressure sensitive adhesive tape No. 18887-48 were labeled and shipped to the NASA Lyndon B. Johnson Space Center.

TABLE VI. TAPE EVALUATIONS

Tape Properties	Initial Contract Specifications	ADL No. 18887-48 Tape
Adhesive Strength on Aluminum (180° Peel)	107-143 grams/cm (0.6-0.8 lb/in)	125-161 grams/cm (0.7-0.9 lb/in)*
Adhesive Strength on Stainless Steel (180° Peel)	71-107 grams/cm (0.4-0.6 lb/in)	107-143 grams/cm (0.6-0.8 lb/in)*
Unwind Resistance	536-714 grams/cm (3-4 lb/in)	680-907 grams (1.5-2.0 lb)***
Tensile Strength	Minimum of 7143 grams/cm (40 lb/in)	7500 grams/cm (42 lb/in)**
Elongation at Break	5-10%	7-8%**
Tear Strength, Elmendorf	200-350 grams (0.44-0.77 lb)	--
Tear Strength, Tongue	363-408 grams (0.8-0.9 lb)	318-408 grams (0.7-0.9 lb) (Machine Direction)
Color	Light	Light (off-white)
Printability	Printable with Pencil or Ball-Point Pen	Printable with Pencil <u>and</u> Ball-Point Pen
Flammability	Self-extinguishing after bottom ignition in 23.8% oxygen, 76.2% nitrogen, 1019 grams/sq. cm (14.5 psia).	Self-extinguishing after bottom ignition in 24% oxygen, 76% nitrogen, atmospheric pressure.

* Tested at 30.5 cm/min (12 in/min) on 2.5 centimeter wide (1 inch wide) specimen.

** Tested at 30.5 cm/min (12 in/min) on 5.1 centimeter wide (2 inch wide) specimen with initial clamp separation of 15.2 centimeters (6 inches).

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