

REPLACED SHEETS		SG
☒	Description : paged 1 to paged 16	
☒	Claim(s) : paged 17 to paged 22	
☐	Drawing(s) : paged to paged	
☐	Abstract : paged to paged	
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THREE-LAYER POLYIMIDESILOXANE ADHESIVE TAPE

Background of the Invention

5 This invention relates to an adhesive tape having a polyimidesiloxane layer on either side of a carrier film. In particular, it relates to a three-layer tape wherein a polyimide carrier film is sandwiched between two polyimidesiloxane adhesive layers.

10 In the electronics industry adhesive tapes are used for a variety of purposes such as, for example, bonding a metal lead frame to an integrated circuit chip. Such tapes, useful for LOC (lead on chip) attachments, are required to be of high purity, have excellent adhesive properties, and be easy to apply using mass production techniques. Where
15 wire bonding is done after the LOC attachment, it is desirable for the polyimidesiloxane adhesive to have a high Tg, usually above about 150°C. Examples of adhesive polyimides can be found in U.S. Patents 3,740,305 and 4,543,295. Also see U.S. Patent 4,480,009.

Some LOC tapes make use of polyimides that are applied as a polyamic acid layer to a polyimide carrier layer and are then imidized. The imidization of a polyamic acid results in the evolution of water and solvent which can
5 cause blistering and a lack of uniform adhesion. A good adhesive tape for use in the electronics and other industries could be applied with heat and pressure and without the evolution of any volatiles.

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Summary of the Invention

We have discovered a superior adhesive tape which will bond together many different materials using only heat and pressure and without the evolution of volatiles. The adhesive tape of this invention consists of a carrier layer sandwiched between two fully imidized polyimidesiloxane adhesive layers. Because the polyimidesiloxane adhesive layers are fully imidized and are solventless, they do not evolve water, solvent, or other volatiles when heat and pressure are used to bond surfaces to the tape.

In order to manufacture this unusual tape, it is necessary to form a solution of a fully imidized polyimidesiloxane adhesive in a solvent. The solution is then applied to the carrier layer and the solvent is evaporated to form a fully imidized polyimidesiloxane. Surprisingly, a fully imidized polyimidesiloxane adhesive layer formed in this manner from solution without heat and pressure will adhere to the carrier layer as a non-tacky film, yet will flow and form a bond to other surfaces when heat and pressure are applied.

We have also found that tapes prepared according to the process of this invention have advantages over identical tapes made by imidizing a polyamic acid layer on a carrier layer. A major advantage of the tapes of this invention is that they can be made rapidly since it is only necessary to evaporate the solvent from the adhesive layer and it is not necessary to imidize the adhesive layer.

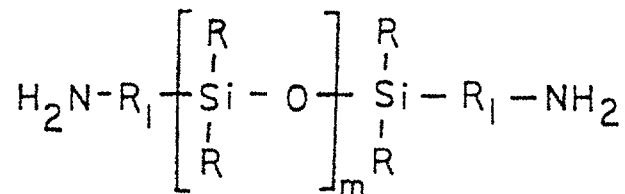


Description of the Invention

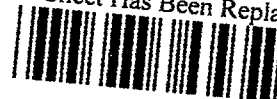
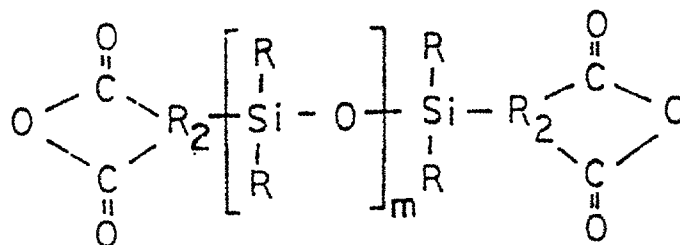
The polyimidesiloxanes used in the process and product of this invention must not only be fully imidized but must also be thermoplastic and soluble in a solvent.

Polyimidesiloxanes are formed by reacting a dianhydride with a diamine, where either a portion of the dianhydride or a portion of the diamine contains siloxane groups. That is, one uses either a dianhydride, a non-siloxane containing diamine, and a siloxane containing diamine, or one uses a dianhydride, a non-siloxane containing diamine, and a siloxane containing dianhydride. It is preferable to use siloxane containing diamines as they are more available and have good properties.

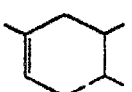
The siloxane-containing compounds may be either aromatic or non-aromatic, but non-aromatic compounds are preferred as they are more readily available. Examples of siloxane diamines that can be used include compounds having the formula



Examples of siloxane dianhydrides that can be used include compounds having the formula



where R, R₁, and R₂ are mono, di, and triradicals, respectively, each independently selected from a substituted or unsubstituted 1 to 12 carbon atom aliphatic group or a substituted or unsubstituted 6 to 10 carbon atom aromatic group. Examples of monoradicals include -CH₃, -CF₃,
 5 -CH=CH₂, -(CH₂)_nCF₃, -C₆H₅, -CF₃-CHF-CF₃, and
 -CH₂-CH₂-C(=O)-O-CH₂CF₂CF₂CF₃. Examples of diradicals include

10 -(CH₂)_n-, -(CH₂)_m-, -CF₂- and -C₆H₄-. Examples of triradicals include -CH-CH₂-,  and 

where n = 1 to 10 and m is 1 to 200, but is preferably 1 to 12. (Siloxane diamines are herein denoted by the notation "G_n".)

15 Any dianhydride may be used in forming the polyimidesiloxane, although aromatic dianhydrides are preferred as they give superior properties. Examples of suitable dianhydrides include

1,2,5,6-naphthalene tetracarboxylic dianhydride;
 20 1,4,5,8-naphthalene tetracarboxylic dianhydride;
 2,3,6,7-naphthalene tetracarboxylic dianhydride;
 2-(3',4'-dicarboxyphenyl) 5,6-dicarboxybenzimidazole
 dianhydride;
 2-(3',4'-dicarboxyphenyl) 5,6-dicarboxybenzoxazole
 25 dianhydride;
 2-(3',4'-dicarboxyphenyl) 5,6-dicarboxybenzothiazole
 dianhydride;
 2,2',3,3'-benzophenone tetracarboxylic dianhydride;



2,3,3',4'-benzophenone tetracarboxylic dianhydride;
3,3',4,4'-benzophenone tetracarboxylic dianhydride (BDTA);
2,2',3,3'-biphenyl tetracarboxylic dianhydride;
2,3,3',4'-biphenyl tetracarboxylic dianhydride;
3,3',4,4'-biphenyl tetracarboxylic dianhydride (BPDA);
bicyclo-[2,2,2]-octen-(7)-2,3,5,6-tetracarboxylic-2,3,5,6-
dianhydride;
thio-diphthalic anhydride;
bis (3,4-dicarboxyphenyl) sulfone dianhydride;
bis (3,4-dicarboxyphenyl) sulfoxide dianhydride;
bis (3,4-dicarboxyphenyl oxadiazole-1,3,4) paraphenylene
dianhydride;
bis (3,4-dicarboxyphenyl) 2,5-oxadiazole 1,3,4-dianhydride;
bis 2,5-(3',4'-dicarboxydiphenylether) 1,3,4-oxadiazole
dianhydride;
bis (3,4-dicarboxyphenyl) ether dianhydride or oxydiphthalic
anhydride (ODPA);
bis (3,4-dicarboxyphenyl) thioether dianhydride;
bisphenol A dianhydride;
bisphenol S dianhydride;
2,2-bis (3,4-dicarboxyphenyl) hexafluoropropane dianhydride
or 5,5-[2,2,2-trifluoro-1-(trifluoromethyl)ethylidene]
bis-1,3-isobenzofurandione) (6FDA);
hydroquinone bisether dianhydride;
bis (3,4-dicarboxyphenyl) methane dianhydride;
cyclopentadienyl tetracarboxylic acid dianhydride;
cyclopentane tetracarboxylic dianhydride;
ethylene tetracarboxylic acid dianhydride;



perylene 3,4,9,10-tetracarboxylic dianhydride;
pyromellitic dianhydride (PMDA);
tetrahydrofuran tetracarboxylic dianhydride; and
resorcinol dianhydride.

5 The anhydrides can be used in their tetraacid form or
as mono, di, tri, or quatra esters of the tetra acid, but
the dianhydride form is preferred because it is more
reactive.

10 The preferred dianhydrides are ODPA, BPDA, BTDA, 6FDA,
and PMDA or mixtures thereof, as these dianhydrides are
readily available and have been found to give superior
properties. The most preferred dianhydride is ODPA because
it gives a polyimidesiloxane having better adhesion and good
flexibility.

15 The non-siloxane containing diamine should be aromatic
as those diamines give the best properties. Examples of
suitable aromatic diamines include
m- and p-phenylenediamine;
2,4-(TDA), 2,5- and 2,6-diaminotoluene;
20 p- and m-xylylenediamine;
4,4'-diaminobiphenyl;
4,4'-diaminodiphenyl ether or 4,4'-oxydianiline
3,4'-oxydianiline (ODA);
4,4'-diaminobenzophenone;
25 3,3', 3,4', or 4,4-diaminophenyl sulfone or m,m-, m,p- or
p,p- sulfone dianiline;
4,4'-diaminodiphenyl sulfide;



3,3' or 4,4'-diaminodiphenylmethane or m,m- or p,p-methylene dianiline;

3,3'-dimethylbenzidine;

4,4'-isopropylidenedianiline;

1,4-bis(p-aminophenoxy)benzene;

1,3-bis(p-aminophenoxy)benzene;

4,4'-bis(4-aminophenoxy)biphenyl;

1,3-bis(3-aminophenoxy)benzene (APB);

2,4-diamine-5-chlorotoluene;

2,4-diamine-6-chlorotoluene;

2,2-bis-4[4-aminophenoxy]phenyl propane (BAPP);

trifluoromethyl-2,4-diaminobenzene;

trifluoromethyl-3,5-diaminobenzene;

2,2'-bis(4-aminophenyl)-hexafluoropropane;

2,2-bis(4-phenoxy aniline) isopropylidene;

2,4,6-trimethyl-1,3-diaminobenzene;

4,4'-diamino-2,2'-trifluoromethyl diphenyloxide;

3,3'-diamino-5,5'-trifluoromethyl diphenyloxide;

4,4'-tri-fluoromethyl-2,2'-diamino biphenyl;

2,4,6-trimethyl-1,3-diaminobenzene;

diaminoanthraquinone;

4,4'-oxybis[2-trifluoromethyl)benzenamine] (1,2,4-OBABTF);

4,4'-oxybis[(3-trifluoromethyl)benzenamine];

4,4'-thiobis[(2-trifluoromethyl)benzenamine];

4,4'-thiobis[(3-trifluoromethyl)benzenamine];

4,4'-sulfoxylbis[(2-trifluoromethyl)benzenamine];

4,4'-sulfoxylbis[(3-trifluoromethyl)benzenamine];

4,4'-ketobis[(2-trifluoromethyl)benzenamine];



4,4'-[(2,2,2-trifluoromethyl-1-(trifluoromethyl)-
ethylidene)bis(3-trifluoromethyl)benzenamine];
4,4'-dimethylsilylbis[(3-trifluoromethyl)benzenamine].

The preferred aromatic diamines are TDA, APB, and BAPP
5 due to their excellent properties.

The polyimidesiloxane can be made from about 1 to about
80 wt% siloxane-containing monomers and about 20 to about 99
wt% monomers that do not contain siloxane. Preferably, it
is made from about 1 to about 30 wt% siloxane-containing
10 monomers and about 70 to about 99 wt% monomers that do not
contain siloxane. Generally, stoichiometric quantities of
diamine and dianhydride are used to obtain the highest
molecular weight polyimidesiloxane but the equivalent ratio
of dianhydride to diamine can range from 1:2 to 2:1.

15 The polyimidesiloxanes are typically prepared in
solution. The solvent used to form the solution of the
polyimidesiloxane must, of course, dissolve the
polyimidesiloxane. Suitable solvents depend upon the
particular composition of the polyimidesiloxane that is to
20 be made and dissolved, but may include N-methylpyrrolidinone
(NMP), diglyme, triglyme, cyclohexanone, cyclopentanone,
dimethylacetamide, and mixtures of these solvents. The
solvent preferably has a boiling point between 130 and 210°C
as lower boiling solvents may evaporate too readily from the
25 completed tape and higher boiling solvents may be too
difficult to remove from the tape. The solution of the
polyimidesiloxane in the solvent can be any percent solids
desired, but it is preferably about 10 to about 30 wt%



solids as more dilute solutions mean more solvent to evaporate and more concentrated solutions are too viscous. A particularly preferred polyimidesiloxane adhesive is made from ODP, APB, and G₉ in NMP.

The first reaction, which forms the polyamic acid, occurs at room temperature and the second reaction, which closes the imide ring, occurs at a temperature of about 150 to about 180°C. Typically, the reaction mixture is refluxed several hours to imidize the polymer. A more complete description of the preparation of solvent-soluble fully imidized thermoplastic adhesive polyimidesiloxanes can be found in U.S. Patent 4,973,645, herein incorporated by reference.

The carrier film can be made from almost any organic polymeric material to which the polyimidesiloxane can adhere. The polymeric material should have sufficient solvent resistance to the polyimidesiloxane solution to prevent its dissolution. The surface of the carrier film can be prepared to enhance the adhesion of the polyimidesiloxane to it. Such preparation may include treatment with a corona or with various chemicals. Many types of carrier films are sold pretreated to increase their adhesion. Preferably, the carrier film is either an amorphous polymer with a T_g greater than 150°C or a crystalline polymer with a melting point greater than 150°C so that it can easily withstand the bonding conditions. Examples of such carrier films include polyimides, polyimidesiloxanes, polyethylene terephthalate,



polyetheretherketones, polysulfones, and polyamides, such as nylon 66. The preferred material for the carrier film is a polyimide. The thickness of the carrier film can vary from about 1/2 to about 10 mils, but it is preferably about 1 to about 2 mils thick.

The tapes of this invention are made by applying the solution of the polyimidesiloxane to the carrier film and evaporating the solvent from the solution. Typically, the carrier film is positioned horizontally while the solution is spread over its upper surface with a doctor blade. The solvent is then evaporated, the carrier film is inverted, and the procedure is repeated. This can be accomplished by hand or in an automated process. Other methods of manufacturing the tape, such as by dipping or running the carrier film through a solution of the polyimidesiloxane followed by wiping and evaporation, are also contemplated. Repeated applications of the solution may be required to build up the desired thickness of adhesive on the tape. The adhesive layer can have a thickness of about 0.1 to about 5 mils on each side of the carrier film and a preferred thickness is about 0.5 to about 1 mils on each side.

After the tape has been made and the solvent evaporated, the adhesive surfaces are no longer tacky and the tape can be rolled up and stored for use. During use the tape is cut to the desired size and the surfaces to be bonded to it are heated and pressed against it. Typically, a temperature of about 200 to about 350°C is used and a pressure of about 10 to about 200 psi. The tape forms a



bond quickly and generally the pressure needs to be applied for only about 1 to about 30 seconds.

Although the tape can be used for many different applications, the principal use contemplated for the tape of this invention is to bond components in the microelectronics industry. Such uses include bonding metal lead frames to integrated circuit chips, chips to lead frames, lead frames to each other, chips to chip carriers and chips to heat spreaders.

The following examples further illustrate this invention.

Example 1

To a dry 12 liter 3 neck flask equipped with overhead stirrer, thermometer, Dean-Stark trap, and nitrogen inlet on top of the condenser was added 5 liters dry NMP and 750 milliliters of toluene. To this flask, with stirring, was added 503.64 grams (1.62 moles) ODPA and then 24 grams (.096 mole) G₁ and 96 grams (.114 mole of siloxane diamine with average molecular weight = 841) G₉. The reaction was stirred four hours and then 577.92 grams (1.41 moles) BAPP was added. The reaction was stirred overnight and the next day, 250 milliliters of NMP and 18 grams DABCO was added. The reaction was heated to reflux and water removed. The reflux was maintained for 4 hours with the pot temperature gradually increasing from about 155°C to about 172°C. About 120 cc of an aqueous phase was removed during this period. After this time, vacuum was applied to remove about 35 - 40% of the solvent. The reaction mixture was cooled to about



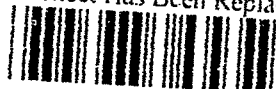
80°C and precipitated into water. The mixture was filtered, washed, reslurried in deionized water and refiltered. The precipitate was then dried at 105°C for three days.

Examples 2 - 7

5 Example 1 was repeated using different monomers. The following table summarizes the preparation of these polyimidesiloxane adhesives:

10 The Glass Transition Temperature T_g was determined by Dynamic Mechanical Thermal Analysis (DMTA). The T_g is taken as the temperature at which the dissipation Tan delta peaks at a heating rate of 4C/min in tension mode at 1 Hz.

Exam ple	Dianhydride (pbw)	Diamine (pbw)	Siloxane Diamine (pbw)	T _g °C
15 1	503.64 ODPA	577.92 BAPP	24G ₁ + 96 G ₉ (MW=841)	208
2	490.8 ODPA	610.4 BAPP	100.28 G ₁₃ (MW=1108)	227
3	71.95 ODPA + 34.42 6FDA	28.36 TDA 23.65 APB	67.73 G ₉ (MW=841) 37.5 G ₉ (MW=841)	215 124
4	38.93 ODPA	63.91 TDA	194.93 G ₉ (MW=867)	185
20 5	241.16 BTDA	40.8 APB	1.0G ₁ + 10.0 G ₉ (MW=841)	187
6	45.7 BPDA	21.64 TDA	311.21 G ₉ (MW=860)	62
7	167.15 ODPA			



Example 8

The polyimidesiloxane of Example 1 was heated in nitrogen at 190°C for 15 hours to increase the molecular weight and then dissolved in NMP and the solution filtered through a 5 micron cartridge filter to give about 3.5 liters of a solution having 21% by weight solids and a viscosity of 95 poise. This solution was further diluted with about 100 cc of NMP to decrease viscosity and then used to coat Upilex SGA polyimide (from Ube). This polyimide was used in a 12" width and 2 mil thickness in roll form. The polyimide sheet was passed through a two roll coater equipped with a trough containing the polyimidesiloxane solution and a knife to control thickness and then through a two zone heated oven with zone 1 (forty feet length) set at 93°C and zone 2 (one hundred and ten feet length) set at 177°C. The polyimide was coated at a rate of 25 feet per minute. After the first side was coated, the single sided sheet was passed through the coater a second time under the same conditions to give a polyimide coated on each side with the polyimidesiloxane of Example 1. The total thickness of the sheet was 3.6 mils.

Example 9

The polyimidesiloxane of Example 2 was dissolved in NMP and the solution filtered through a 5 micron cartridge filter to give about 3.5 liters of a solution having 16% by weight solids and a viscosity of 9.0 poise. This polyimidesiloxane was used as in Example 8 to give a polyimide coated on each side with the polyimidesiloxane of Example 2. The total thickness of the sheet was 3.6 mils.



Example 10

The polyimidesiloxane of Example 3 was heated at 200°C for 19 hours to increase the molecular weight and then dissolved in NMP and the solution filtered through a 5 micron cartridge filter to give about 3.5 liters of a solution having 24% by weight solids and a viscosity of 7.5 poise. This polyimidesiloxane was used as in Example 8 to give a polyimide coated on each side with the polyimidesiloxane of Example 3. The total thickness of the sheet was 3.6 mils.

Example 11

The polyimidesiloxane of Example 4 was dissolved in cyclohexanone to give a form a homogeneous but hazy solution containing 25% by weight solids. Strips of PET (polyethyleneterephthalate) film 5 mils thick and about 0.5 inch wide were dipped in the polymer solution then hung up to drain off and air dry for 5 days. The coated film was about 6.3 mils thick.

Example 12

A nylon 6-6 film which is about 4 mils thick was coated as in example 11. The coated film was about 6 mils thick.

Example 13

The pelyimidesiloxane of Example 5 was dissolved in cyclohexanone to give a solution containing 25% by weight solids. A strip of 2 mil thick Kapton H was dipped in the solution and allowed to drain off. The coated strip was then dried in a 150 C oven for about 1 hour. The coated film was about 4 mils thick.



Example 14

The polyimidesiloxane of Example 6 was dissolved in NMP to give a solution containing 25% by weight solids. A strip of 2 mil thick polyimide film (Kapton H from DuPont) was dipped in this solution and allowed to drain off and air dry. It was dried further in a 200°C oven for about 1 hour.

Example 15

Example 14 was repeated except that 1 mil thick Kapton H was used.

Example 16

The polyimidesiloxane of Example 6 was dissolved in diglyme to give a solution containing 40 % by weight solids. The solution was filtered through a 10 micron filter and then a 1 micron filter. Strips of Kapton H, 2 5/16 inches wide and 2 mils thick and about 6 inches long were passed through the above solution and hung up to air dry for several days.

Example 17

A strip (about 1 cm²) was cut from the coated tape of Example 8 and placed between a strip of alloy 42 metal and the coated side of a piece of silicon wafer precoated with a PMDA-ODA polymer. (This precoating was done by spinning a solution of 0.17 % 3-aminopropyltriethoxysilane on a 4 inch silicon wafer for 30 seconds at 5000 rpm, then a 15 % solution of PMDA- 4,4'-ODA polyamic acid in 75/25 NMP/xylene for 30 seconds at 3000 rpm and drying at 5° C/minute to 350° C/60 minute in a nitrogen atmosphere). The assembly was placed on a hot plate at 275°C and with no pressure for 10



seconds. Then 5.5 pounds of pressure was applied for 10 seconds and the assembly was removed and the adhesion tested on a HMP model 1750 die shear tester. Four assemblies were tested and averaged to give a shear adhesion of greater than 25 kg/cm².

Examples 18 - 25

Example 17 was repeated using the tape of Examples 9 to 16. The following table gives the results:

	Example	Tape from Example	Hot Plate Temperature (°C)	Shear Adhesion (kg/cm ²)
10	17	8	275	>25
	18	9	350	>25
	19	10	275	>25
	20	11	225	>25
	21	12	225	>25
15	22	13	275	>25
	22A	13(A) ¹	275	>25
	22B	13(B) ²	275	>25
	23	14	275	>25
	24	15	275	>25
20	25	16	250	>25

¹ Same as Example 13, but between copper and an uncoated silicon wafer.

² Same as Example 13, but between ceramic and an uncoated silicon wafer.

These examples demonstrate that the trilayer films of this invention can be used to form strong bonds between various surfaces.





WE CLAIM:

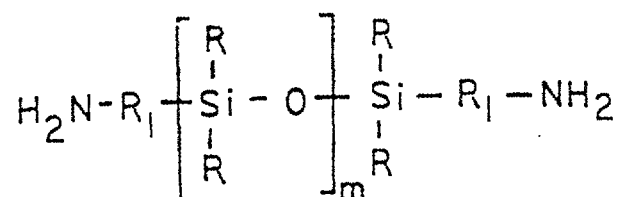
1. An adhesive tape comprising a carrier film sandwiched between two layers of fully imidized thermoplastic polyimidesiloxane adhesive.
2. An adhesive tape according to Claim 1 wherein said carrier film is selected from polyimide, polyimidesiloxane, polyethylene terephthalate, and polyamide.
3. An adhesive tape according to Claim 1 wherein said carrier film is polyimide.
4. An adhesive tape according to Claim 1 wherein said carrier film is about 1/2 to about 10 mils. thick.
5. An adhesive tape according to Claim 1 wherein said polyimidesiloxane adhesive comprises the reaction product of an aromatic dianhydride, a non-siloxane containing diamine, and siloxane containing diamine.
6. An adhesive tape according to Claim 5 wherein said siloxane diamine comprises about 1 to about 70 wt% of said polyimidesiloxane.
7. An adhesive tape according to Claim 5 wherein said aromatic dianhydride is selected from the group consisting of benzophenone tetracarboxylic acid dianhydride, oxydiphthalic anhydride, biphenyl dianhydride, 6FDA, pyromellitic dianhydride, and mixtures thereof.





8. An adhesive tape according to Claim 7 wherein said aromatic dianhydride is oxydiphthalic anhydride.
9. An adhesive tape according to Claim 5 wherein said aromatic diamine is selected from the group consisting of toluene diamine, 1,3-bis(3-aminophenoxy)benzene, and 2,2-bis(4[4-aminophenoxy]phenyl)propane.

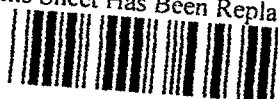
10. An adhesive tape according to Claim 5 wherein said siloxane diamine has the formula



where R is a monovalent radical and R₁ is a divalent radical and each R and R₁ are independently selected from substituted or unsubstituted 1 to 12 carbon atom aliphatic group and substituted and unsubstituted 6 to 10 carbon atom aromatic group and m is 1 to 200.

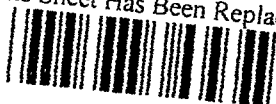
11. An adhesive tape according to Claim 10 wherein said aromatic dianhydride is oxydiphthalic anhydride, said non-siloxane containing diamine is 1,3-bis(3-aminophenoxy)benzene, "m" in the formula for said siloxane diamine is 9, and said solvent is N-methylpyrrolidinone.

12. A method making an adhesive tape comprising
- A. forming a solution in a solvent of a fully imidized polyimidesiloxane;
 - B. applying said solution to both surfaces of a carrier film; and





- C. evaporating the solvent from said solution.
13. A method according to Claim 12 wherein said solution is applied first to one surface of said carrier film and the solvent is evaporated and then said solution is applied to the other surface of said film and then the solvent is evaporated.
14. A method according to Claim 12 wherein said solution of polyimidesiloxane is formed by dissolving in said solvent an aromatic dianhydride, a non-siloxane containing diamine, and a siloxane containing diamine, heating said solution to form a polyamic acid, and further heating said solution to form a polyimidesiloxane.
15. A method according to Claim 12 wherein the boiling point of said solvent is about 130 to about 210°C.
16. A method according to Claim 12 wherein said solution is about 10 to about 30 wt% solids.
17. A method according to Claim 12 including the additional last step of placing said adhesive tape between two surfaces and pressing two said surfaces together at a temperature of about 200 to about 350°C at pressure of about 10 to about 200 psi for about 1 to about 30 seconds.
18. A method according to Claim 17 wherein said surfaces are a metal lead frame and an integrated circuit chip.
19. A method according to Claim 12 including the additional last step of rolling said tape up.



20. A rolled up tape made according to the method of Claim
19.



This Sheet Has Been Replaced

