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ADHESIVE FILMBackground of the Invention

5

Field of the Invention

The present invention relates to a cyanate ester adhesive film which can be cured extremely rapidly and has excellent storage stability.

10

Description of the Related Art

Various types of adhesives are known in the art. For use in the electronics industry, an adhesive must meet specific requirements. First, in the swiftly growing semiconductor industry, a rapid cure rate, e.g., 15 30 seconds or less at 180°C, is required for productivity. These conditions, formerly known as "rapid curing" are becoming insufficient for productivity reasons, and even faster curing adhesives are being required. Further, the adhesive used must be unreactive 20 after curing, and noncorrosive to metals. Most conventional adhesives cannot meet these strict criteria.

Phenol resin/nitrile rubber thermally curable adhesives are well known. However, due to the use of phenol as the thermal curing agent, the phenol 25 resin/nitrile rubber adhesives are slow curing, and are extremely corrosive to metals. Further, they may pose a safety risk by containing unreacted formaldehydes. The emulsion-polymerized nitrile rubber thermoplastic ingredient, is thus unsuitable for electronic 30 applications, due to possible corrosion of the electronic article and/or malfunction of the semiconductor.

Urethane/isocyanate adhesives are also well known in the adhesive art. However, the urethane resin/isocyanate adhesives have poor storage stability, and inferior 35 mechanical strength and heat resistance.

Epoxy resin/PVB adhesives also cure too slowly for the desired applications, and may contain undesired halogens generated from the preparation.



-2-

Various "improved" adhesives have been proposed for use in the electronics industry.

5 An adhesive having improved curing time is disclosed in JP 52-22849, comprising a three-layered film in which an epoxy resin and a curing agent are divided into the outside layers with a diaphragm layer therebetween, and an unsaturated compound, e.g., an acrylic monomer, and a peroxide curing catalyst are similarly divided in order to improve reactivity.

10 However, peroxide is extremely dangerous to handle and store, especially in the presence of metal particles, and unsaturated acrylic monomers have odor and toxicity problems. Further, even when carefully formulated, the epoxy resin and the curing agent may not mix exactly as
15 desired, causing variation in the adhesive film. Also, the method of preparation is expensive, and requires many complicated steps.

Another rapidly curing adhesive, a combination of a cyanate ester resin with an organometallic compound, is
20 disclosed in JA 2-105823. The adhesive resin has relatively good storage stability, however the metal catalyst may affect the release agent after processing into a film.

In the above adhesives, the cure rate could only
25 been changed by varying the type and/or amount of the curing catalyst. When adhesives having different curing rates are required, it is necessary to prepare each adhesive separately.

~ An anisotropic conductive membrane in which
30 conductive particles are added to an adhesive film is also known for use with electronics. However, the thickness must be selected for the substance to be adhered. In the conventional technique, it is necessary to prepare anisotropic conductive adhesive membranes for
35 each substance to be adhered.

It has now been discovered that an adhesive comprising a primary agent layer containing a cyanate



-3-

ester resin and a thermoplastic resin, and a catalyst layer comprising at least one organometallic compound, metal chelate or organic metal salt, and a thermoplastic resin can be cured very rapidly, has excellent storage stability, and is not corrosive to metal.

Summary of the Invention

The present invention provides an adhesive film which can be cured very rapidly, and which has excellent storage stability.

Specifically, adhesive films of the present invention comprise an primary agent layer cyanate ester resin and a thermoplastic resin, and a catalyst layer comprising at least one organometallic compound, metal chelate or organic metal salt, and a thermoplastic resin.

Preferred adhesive films comprises

a) at least one primary agent layer comprising a cyanate ester resin having a molecular weight of from about 150 to about 2,000, and a thermoplastic resin having a molecular weight of from about 3,000 to about 200,000, and

b) at least one catalyst layer comprising from about 0.01 to about 10 parts by weight of at least one curing agent selected from the group consisting of organometallic compounds, metal chelates, and organic metal salts, and a thermoplastic resin having a molecular weight of from about 3,000 to about 200,000, wherein the sum of the thermoplastic resin in both the primary agent layer and the catalyst layer is from about 10 to about 300 parts per 100 parts cyanate ester resin.

Detailed Description of the Invention

The cyanate ester resin is suited for use in the electronic industry. The thermal resistance and creep resistance are excellent, and the resin may be cured very quickly. The resins are also self-crosslinking.

Useful cyanate ester resins contain monomer and

-4-

oligomers having at least one cyanate group in the molecular chain and derivatives thereof.

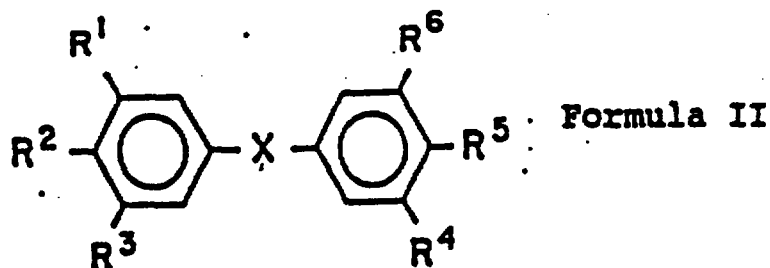
Preferred compositions of the invention include one or more cyanate esters according to Formulas I, II, III or IV. Formula I is represented by



Formula I

wherein p can be from 2 to 7, and wherein Q comprises at least one of the following categories: (1) a mono-, di-, tri-, or tetra-substituted aromatic hydrocarbon containing from about 5 to about 30 carbon atoms, (2) a 1 to 5 aliphatic or polycyclic aliphatic mono-, di-, tri- or tetra-substituted hydrocarbon containing from about 7 to about 20 carbon atoms. Optionally, either category may contain from about 1 to about 10 heteroatoms selected from the group consisting of non-peroxidic oxygen, sulfur, non-phosphino phosphorous, non-amino nitrogen, halogen, and silicon.

Formula II is represented by



where X is a single bond, a lower alkylene group having from 1 to 4 carbons, -S-, or the SO₂ group; and where R¹, R², R³, R⁴, R⁵, and R⁶ are independently hydrogen, an alkyl group having from one to three carbon atoms, or the cyanate ester group (-OC≡N), with the proviso that at least two of R¹, R², R³, R⁴, R⁵, and R⁶ are cyanate ester groups. In preferred compounds, each of the R groups is either -H, methyl or the cyanate ester group.

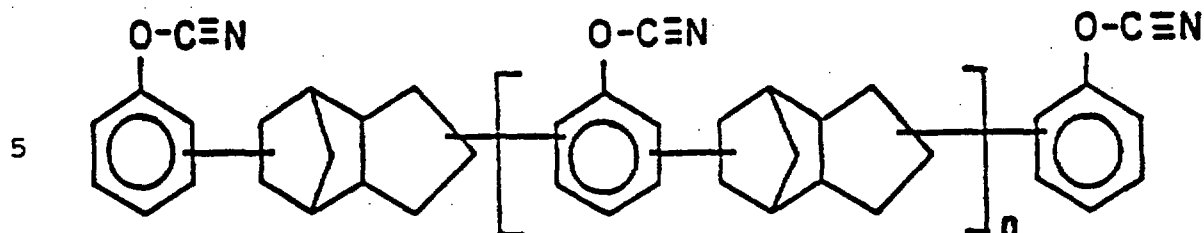
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-5-

Formula III is represented by

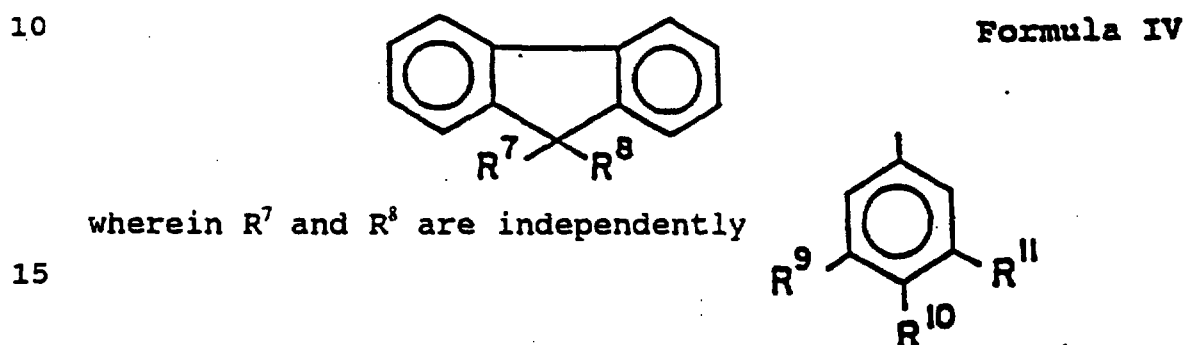
Formula III



where n is from 0 to about 5.

Formula IV is represented by

Formula IV

wherein R^7 and R^8 are independently

wherein R^9 , R^{10} , R^{11} are independently -H, a lower alkyl group having from about 1 to about 5 carbon atoms, or the cyanate ester group, preferably hydrogen, methyl or the cyanate ester group, with the proviso that R^7 , and R^8 combined include at least two cyanate ester groups.

Useful cyanate ester compounds include, but are not limited to the following:

- 1,3- and 1,4-dicyanatobenzene;
- 2-tert-butyl-1,4-dicyanatobenzene;
- 2,4-dimethyl-1,3-dicyanatobenzene;
- 2,5-di-tert-butyl-1,4-dicyanatobenzene;
- tetramethyl-1,4-dicyanatobenzene;
- 4-chloro-1,3-dicyanatobenzene;
- 1,3,5-tricyanatobenzene;
- 2,2'- and 4,4'-dicyanobiphenyl;
- 3,3',5,5'-tetramethyl-4,4'-dicyanobiphenyl;
- 1,3-, 1,4-, 1,5-, 1,6-, 1,8-, 2,6-, and 2,7-
- dicyanatonaphthalene;
- 1,3,6-tricyanatonaphthalene;
- bis(4-cyanatophenyl)methane;

This Sheet Has Been Replaced





- bis(3-chloro-4-cyanatophenyl)methane;
bis(3,5-dimethyl-4-cyanatophenyl)methane;
1,1-bis(4-cyanatophenyl)ethane;
2,2-bis(4-cyanatophenyl)propane;
5 2,2-bis(3,3-dibromo-4-cyanatophenyl)propane;
2,2-bis(4-cyanatophenyl)-1,1,1,3,3,3-hexafluoropropane;
bis(4-cyanatophenyl)ester;
bis(4-cyanatophenoxy)benzene;
bis(4-cyanatophenyl)ketone;
10 bis(4-cyanatophenyl)thioether;
bis(4-cyanatophenyl)sulfone;
tris(4-cyanatophenyl)phosphate, and
tris(4-cyanatophenyl)phosphate.

- Also useful are cyanic acid esters derived from
15 phenolic resins, e.g., as disclosed in U.S. Patent
3,963,184, cyanated novolac resins derived from novolac,
e.g., as disclosed in U.S. Patent No. 4,022,755, cyanated
bis-phenol-type polycarbonate oligomers derived from
bisphenol-type polycarbonate oligomers, as disclosed in
20 U.S. Patent 4,096,913, cyano-terminated polyarylene
ethers as disclosed in U.S. Patent No. 3,595,000, and
dicyanate esters free of ortho hydrogen atoms as
disclosed in U.S. Patent No. 4,740,584, mixtures of di-
and tricyanates as disclosed in U.S. Patent No.
25 4,709,008, polyaromatic cyanates containing polycyclic
aliphatic groups as disclosed in U.S. Patent No.
4,528,266, e.g., QUARTEX™ 7187, available from Dow
Chemical, fluorocarbon cyanates as disclosed in U.S.
Patent No. 3,733,349, and cyanates disclosed in U.S.
30 Patents 4,195,132, and 4,116,946, all of the foregoing
patents being incorporated by reference.

Polycyanate compounds obtained by reacting a phenol-
formaldehyde precondensate with a halogenated cyanide are
also useful.

- 35 Examples of preferred cyanate ester compositions
include low molecular weight oligomers, e.g., from about
250 to about 1200, of bisphenol-A dicyanates such as



-7-

AroCy UC-30 Cyanate Ester Semisolid Resin, commercially available from Hi-Tex polymers, Jeffersontown, Kentucky; low molecular weight oligomers of tetra o-methyl bisphenol F dicyanates, such as AroCy M-30 Cyanate Ester Semisolid Resin, also commercially available from Hi-Tek Polymers; low molecular weight oligomers of thiodiphenol dicyanated, such as AroCy T-30 Cy.

Useful thermoplastic resins are those having good compatibility with cyanate ester, and film-forming properties to aid in flexibility. Included in this class are polyvinylformyls, polyvinylacetals, polyvinylbutylols, polyamide, polyimides, polysulfones, polyesters, polyesterimides, polycarbonates, polycarbonatesiloxanes, polyethers, polyallylates, polyimidesiloxanes, phenoxy resins, carboxylated SEBS, acrylic resins, and polyurethanes. Useful thermoplastic resins are those having molecular weights between about 3,000 and about 200,000. If the molecular weight is below about 3000, the resin has a poor film-forming ability, and the resulting film is brittle; if the molecular weight is over 200,000, the thermoplastic and the cyanate ester will not mix adequately, and the curing time is significantly extended.

Useful thermoplastic resins are similar for those in the catalyst layer and those in the primary agent layer. The thermoplastic resin used in the two layers may be the same or different.

The amounts of thermoplastic resin in the primary agent layer and in the catalyst layer together comprise from about 10 to about 300 parts to 100 parts of cyanate ester resin.

Known curing catalysts are useful in compositions of the invention. An organometallic compound, metal chelate or an organometallic salt are suitable candidates. Examples of organometallic compounds, such as acetylacetonate copper and cobalt acrylacetonate are disclosed in JP 2-105823, incorporated herein by



-8-

reference. Examples of the organometallic salts include cobalt naphthenate and copper anthenate. These catalysts are activated by both heat and light, and therefore exposure to heat can shorten the curing time even more.

- 5 They are easily dissolved and dispersed in the adhesive solution.

Catalysts for the reaction of the cyanate ester include organometallic compounds containing a cyclopentadienyl group, C_5H_5 , and suitable derivatives, such as cyclopentadienyl iron dicarbonyl dimer, $[C_5H_5Fe(CO)_2]_2$, pentamethylcyclopentadienyl iron dicarbonyl dimer, $[C_5(CH_3)_5Fe(CO)_2]_2$, methylcyclopentadienyl manganesetricarbonyl, all available from Strem Chemical Company, Newburyport, Massachusetts, and hexafluorophosphate salt of the cyclopentadienyl iron mesitylene cation, C_5H_5 (mesitylene) $Fe^+PF_6^-$, and trifluoromethanesulfonate salt of the cyclopentadienyl iron mesitylene cation, C_5H_5 (mesitylene) $Fe^+CF_3SO_3^-$, both of which may be prepared as described in U.S. Patent 4,868,277, incorporated herein by reference.

Other organometallic compounds which are suitable catalysts for use in the present invention are disclosed in EPO Publication No. 364,073.

Suitable sources of radiation for photoactivation of the catalysts include such conventional sources as mercury vapor discharge lamps, tungsten lamps, xenon lamps, fluorescent lamps, sunlamps, lasers, carbon arcs, and sunlight. Photoactivated catalysts are preferred as they increase shelf life. The layers are typically formed on a release liner or paper.

The amount of catalyst used preferably falls in the range of from about 0.01 to about 10 parts by weight per 100 parts of the cyanate ester resin.

The number of layers is not critical. Preferred embodiments of the invention contain from about 1 to about 3 catalyst layers, and 1 or 2 primary agent layers. It is important to note, that the catalyst layers are not



-9-

necessarily in contact with the entire surface of the primary agent layer, and may be present on isolated areas of the primary agent layer, by such techniques as screen printing.

5 Thickness of the layers are dependent on such factors as the number of layers and the specific application. The primary agent layer ranges from about 1 micrometer to about 100 micrometers. The catalyst layer ranges from about 0.1 micrometer to about 50 micrometers.

10 If desirable, a third layer may also be present. This layer may be provided in order to improve storage stability, film formation and the like. For example, a polyvinyl butryal, polysulfone, nylon, epoxy/amine or the like can be used as a third layer having a thickness of
15 from about 0.1 micrometer to about 50 micrometers.

Conventional adjuvants such as electrically conductive particles, thermally conductive particles, silane coupling agents, antioxidants, copper deterioration proofing agents, and tackifiers may be
20 added to the adhesive films of the present invention when desired.

Examples of the processes useful for preparation of the adhesive film of the present invention include, but are not limited to, conventional processes such as Meyer
25 bar coating, knife coating, die coating, extrusion, lamination processes, printing, and the like.

As solvents for use in the film formation, methyl ethyl ketone (MEK), tetrahydrofuran (THF), chloroform and the like are useful.

30 The invention can further comprise other thermosetting resins such as phenolic and silicone resins, epoxy hardeners, tackifiers, coupling agents, antioxidants, modifiers and additives without departing from the spirit of the invention.

35 All percents, parts and ratios herein are by weight unless specifically stated otherwise. The following examples are meant to be illustrative and do not limit

-10-

the scope of the invention, which is defined solely by the claims.

Examples

Example 1

5

(1) Formation of the Main Agent Layer:

As a cyanate ester resin, 100 g of AROCY (2,2-bis(4-cyanatophenyl)propane; molecular weight; about 270; supplied from Hi-Tek Polymers) was dissolved in 200 g of methyl ethyl ketone. In this was dissolved 100 g of S-LEC B BX-1 (polyvinyl butyral, molecular weight; about 100,000; supplied from Sekisui Chemical) as a thermoplastic resin to prepare a main agent solution. This was coated on a silicone-treated polyester film having a thickness of 50 μm by a hand coater, and then dried to form a main agent layer having a thickness of 20 μm .

(2) Formation of Curing Catalyst Layer:

As a thermoplastic resin, 100 g of BX-1 was dissolved in 500 g of MEK, and 5 g of cyclopentadienyl iron dicarbonyl dimer ($[\text{C}_5\text{H}_5\text{Fe}(\text{CO})_2]_2$; hereinafter abbreviated as IDC) was added thereto to prepare a homogenous curing catalyst solution. This was coated on a silicone-treated polyester film by a hand coater, and then dried to form a curing catalyst layer having a thickness of 5 μm .

(3) Preparation of the Adhesive Film:

The above-mentioned main agent layer and the curing catalyst layer were laminated by a laminator under the conditions of 100°C x 5 kg/cm² x 2 seconds to obtain an adhesive film.

(4) Evaluation of the Film Formation:

Film formation was evaluated from handling properties and appearance.

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-11-

Very Good: Possible to handle, flexible
Good: Possible to handle, slightly brittle
Normal: Possible to handle, cracks appeared
Poor: Impossible to handle, cracks throughout

5

(5) Evaluation of the Reactivity 1:

The sheet is heated up by means of DSC 4
(differential scanning calorimeter, supplied from Perkin
Elmer) at a rate of 10°C/min. to measure an exothermic
10 peak temperature at this time.

Very Good: Peak Temp. of 150°C or less
Good: Peak Temp. of 170°C or less
Moderate: Peak Temp. of 190°C or less
Poor: Peak Temp. of more than 210°C

15

(6) Evaluation of the Reactivity 2:

Tack free times (TFT) were determined at 180°C by a
plate gel timer.

Very Good: TFT of 10 sec. or less
20 Good: TFT of 30 sec. or less
Moderate: TFT of 60 sec. or less
Poor: TFT of more than 60 sec.

(7) Evaluation of the Storage Stability:

25 A Sample was left standing at either room
temperature or 80°C, and the storage stability was
evaluated by a bending test. In the bending test, the
sample was bent 180°, and the time required for cracking
was measured.

30

Time for Cracking

	<u>Room Temp.</u>	<u>80°C</u>
Very Good:	1 month or more	1 hour or more
Good:	2 weeks or more	30 minutes or more
35 Moderate:	1 week or more	20 minutes or more
Poor:	within 1 week	10 minutes

This Sheet Has Been Replaced



-12-

(8) Evaluation of the Adhesive Force:

An Au plated 35 μ Cu/50 μ polyimide flexible printed circuit (space 0.1 mm/pitch 0.2 mm) and an ITO glass (plane resistance: 30 $\Omega/\square$) were rolled by means of a J3
5 bonder (thermal roller, supplied from Sumitomo 3M) under the conditions of 180°C x 30 kg/cm² x 20 seconds, and a peeling strength at 90° was measured by using a tension at a stress rate of 50 mm/min.

10 Adhering Force

Very Good: 1000 g/cm or more
Good: 500 g/cm or more
Moderate: 300 g/cm or more
Poor: 300 g/cm or more

15

Examples 2-13

In these Examples, types of the cyanate resins, types of thermoplastic resins (including the case in combination with thermosetting resins), and the amount
20 thereof, and types and amounts of curing catalysts were varied. The preparation and evaluation of the films were as in Example 1. The resins and the curing catalysts used are as follows:

25 Cyanate Ester Resin

AROXY B30 (2,2-Bis(4-cyanatophenyl)propane;
Molecular weight about 560; supplied from Hi-Tek
Polymers)

AROXY B50 (2,2-Bis(4-cyanatophenyl)propane;
30 Molecular weight about 1,100; supplied from Hi-Tek
Polymers)

AROXY M30 (Bis-3,5-dimethyl(4-cyanatophenyl)methane;
Molecular weight about 490; supplied from Hi-Tek
Polymers)

35 QUATREX XU71787 (Alicyclic aromatic cyanate ester,
supplied from Dow Chemical Co.)

This Sheet Has Been Replaced





-13-

Thermoplastic Resin

Eryther UE 3300 (Polyester Resin; Molecular weight about 8,000; Unitika Ltd.)

UDEL P1700 (Polysulfone; supplied from Amoco)

5 UCAR PKHC (Phenoxy Resin; Molecular weight about 25,000; supplied from UCC)

Tough Tec M1913 (Carboxylated SEBS resin: Molecular weight about 50,000, supplied from Asahi Chemical Industry Co., Ltd.)

10 S-LEC B BXL (PVB; Molecular weight about 18,000; supplied from Sekisui Chemical)

S-LEC BX5 (PVB; Molecular weight about 150,000; supplied from Sekisui Chemical)

15

Thermosetting Resin

Epicoat 828 (DGEBA epoxy; Molecular weight about 380; supplied from Shell Chemicals)

Sumilec PC 1 (Resol phenol; Sumitomo Dules)

20

Curing Catalyst

IDC

Cobalt acetylacacetate (abbreviated as "AcAcCo")

Copper naphthenate (abbreviated as "NpCu")

25 $\text{Fe}_2(\text{CO})_9, \text{Mn}_2\text{CO}_{10}$

EXAMPLES 14-16

In these examples, the numbers of the main agent layers and the curing catalyst layers varied. The preparation and evaluation of the film were as in Example 1.

30

EXAMPLE 17

In this example, to the main agent layer 10 parts by weight of Fine Pearl Au-10S (Au-plated electrically conductive particle supplied from Sumitomo Chemicals) was added based on 100 parts by weight of XU71787 main agent.

35

-14-

The sample for measuring the adhesive force was aged at 85°C for 85 hours, and the resistance was measured by using a multimeter.

5 COMPARATIVE EXAMPLES 1-5

These comparative examples deviate from the scope of the present invention. The preparation and evaluation of the film were as in Example 1.

10 COMPARATIVE EXAMPLE 6

(1) Preparation of the Film:

To the solution prepared in Example 1 (1), 10 parts by weight of IDC was added based on 100 parts by weight of B10, and then homogeneously dispersed and mixed.

15 Thereafter, the mixture was applied on a silicone-treated polyester film by a hand coater and then dried to obtain an adhesive film.

(2) Evaluation:

20 The evaluation was the same as that of Example 1 (4)-(8).

COMPARATIVE EXAMPLE 7

25 This is an example of the conventional adhesive sheet.

(1) Preparation of the Film:

100 g of BX-1 was dissolved in 500 g of MEK. 100 g of Epicoat 828 and 10 g of Curezole 2E4MZ (imidazoles, supplied from Shikoku Chemical) as a curing agent were
30 added thereto to prepare a homogenous curing catalyst solution. This was applied on a silicone-treated polyester film by a hand coater and then dried to obtain an adhesive film.

35 (2) Evaluation:

The evaluation was the same as that of Example 1 (4)-(8).

This Sheet Has Been Replaced



-15-

COMPARATIVE EXAMPLE 8

This example was the same as that of Comparative Example 7, except that 10 parts by weight of Fine Pearl was added in the adhesive based on 100 parts by weight of 5 Epicoat 828. The resistance measurement was carried out as in Example 17 using a sample for measuring the adhesive force.

These results are shown in Tables 1, Table 2, Table 10 3, Table 4, and Table 5.

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TABLE 1

		Ex. 1	Ex. 2	Ex. 3	Ex. 4	Ex. 5	Ex. 6
Main Agent Layer	Cyanate	B10/100	B50/100	B30/100	M30/100	XU71787/100	B30/100
	Thermoplastic Resin	BX1/100	P1700/100	BE3300/100	PKHC/100	M1913/100	BXL/100
	Thickness (μ)	20	20	10	20	20	20
Curing Catalyst Layer	Catalyst	IDC/5	AcAcCo/5	NpCu/5	IDC/5	IDC/5	Fe2(CO)9/0.05
	Thermoplastic Resin	BX1/100	828/100	PC1/100	BXL/100	M1913/100	BXL/100
	Thickness (μ)	5	5	15	5	5	5
Results	Film Formation	Very Good	Good	Very Good	Very Good	Very Good	Very Good
	DSC	Very Good	Very Good	Very Good	Very Good	Very Good	Good
	TFT	Very Good	Very Good	Very Good	Very Good	Very Good	Good
	Storage Stability (Room Temp.)	Very Good	Very Good	Very Good	Very Good	Very Good	Very Good
	Storage Stability (80°C)	Very Good	Very Good	Very Good	Very Good	Very Good	Very Good
	Adhesive Force	Very Good	Very Good	Very Good	Very Good	Very Good	Good



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TABLE 2

TABLE 2

		Ex. 7	Ex. 8	Ex. 9	Ex. 10	Ex. 11	Ex. 12
Main Agent Layer	Cyanate	B30/100	B30/100	B30/100	XU71787/100	XU71787/100	XU717871/100
	Thermoplastic Resin	BXL/100	BX5/50	BX5/50	BX1/50	BX1/50	BX1/50
	Thickness (μ)	20	20	20	20	20	20
Curing Catalyst Layer	Catalyst	Fe ₂ (CO) ₉ /0.5	Mn ₂ (CO) ₁₀ /10	Mn ₂ (CO) ₁₀ /25	IDC/0.05	IDC/10	Mn ₂ (CO) ₁₀ /5
	Thermoplastic Resin	BX1/100	BXL/100	BXL/100	PKHC/100	M1913/100	M1913/100
	Thickness (μ)	5	5	5	5	10	10
Results	Film Formation	Very Good	Very Good	Very Good	Very Good	Very Good	Very Good
	DSC	Very Good	Very Good	Very Good	Very Good	Very Good	Very Good
	TFT	Very Good	Very Good	Very Good	Very Good	Very Good	Very Good
	Storage Stability (Room Temp.)	Very Good	Very Good	Good	Very Good	Very Good	Very Good
	Storage Stability (80°C)	Very Good	Very Good	Good	Very Good	Very Good	Very Good
	Adhesive Force	Very Good	Very Good	Very Good	Very Good	Very Good	Very Good



TABLE 3

		Ex. 13	Ex. 14	Ex. 15	Ex. 16	Ex. 17
Main Agent Layer	Cyanate	B30/100	B30/100	B30/100	B30/100	XU71787/100 (Au10s/10)
	Thermoplastic Resin	BX1/100	PX1/100	BX1/100	BX1/100	BX1/100
	Thickness (μ)	10	30	20	10x2	20
	No. of Layer	1	1	1	2	1
Curing Catalyst Layer	Catalyst	IDC/5	IDC/5	IDC/5	IDC/5	IDC/5
	Thermoplastic Resin	BX1/100	BX1/100	BX1/100	BX1/100	BX1/100
	Thickness (μ)	10	2	2x2	2x3	5
	No. of Layer	1	1	2	3	1
Results	Film Formation	Very Good	Good	Very Good	Very Good	Very Good
	DSC	Very Good	Very Good	Very Good	Very Good	Very Good
	TFT	Very Good	Very Good	Very Good	Very Good	Very Good
	Storage Stability (Room Temp.)	Very Good	Good	Very Good	Good	Very Good
	Storage Stability (80°C)	Very Good	Good	Very Good	Good	Very Good
	Adhesive Force	Very Good	Very Good	Very Good	Very Good	Very Good

TABLE 4



TABLE 4

		Comp. Ex. 1	Comp. Ex. 2	Comp. Ex. 3	Comp. Ex. 4	Comp. Ex. 5
Main Agent Layer	Cyanate	B30/100	UX71787/100	B30/100	B30/100	B30/100
	Thermoplastic Resin	BXL/100	BXL/100	BXL/3	BXL/50	BX1/300
	Thickness (μ)	10	20	40	10	20
	No. of Layer	1	1	1	1	1
Curing Catalyst Layer	Catalyst	IDC/5	Mn2(CO)10/.01	IDC/10	IDC/5	IDC/1
	Thermoplastic Resin	BXL/100	BXL/100	BXL/100	828/400	BX1/100
	Thickness (μ)	10	5	2	10	5
	No. of Layer	1	1	1	1	1
Results	Film Formation	Very Good	Very Good	Poor	Poor	Very Good
	DSC	Very Good	Poor	Very Good	Poor	Normal
	TFT	Very Good	Poor	Very Good	Poor	Poor
	Storage Stability (Room Temp.)	Normal	Very Good	-	-	Very Good
	Storage Stability (80°C)	Poor	Very Good	-	-	Very Good
	Adhesive Force	Very Good	Very Good	-	-	Normal



-20-

Table 5

5	Initial Value	24 hrs.	168 hrs.	500 hrs.	1000 hrs.	
	Ex. 17	12.3	12.5	12.7	12.8	12.9
10	Comp.Ex. 8	32.9	59.2	(7)	(12)	(13)

Unit: Ω

15 The number in parentheses is the number of failed
conduction terminals per 20 terminals.

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-21-



What is Claimed is:

1. An adhesive film comprising
- a) at least one primary agent layer comprising a cyanate ester resin having a molecular weight of from about 150 to about 2,000, and a thermoplastic resin having a molecular weight of from about 3,000 to about 200,000, and
- b). at least one catalyst layer comprising from about 0.01 to about 10 parts by weight of at least one curing agent selected from the group consisting of organometallic compounds, metal chelates, and organic metal salts, and a thermoplastic resin having a molecular weight of from about 3,000 to about 200,000,
- wherein the sum of the thermoplastic resin in both the primary agent layer and the catalyst layer is from about 10 to about 300 parts per 100 parts cyanate ester resin.
2. The adhesive film of claim 1 wherein the primary agent layer and the catalyst layer are in contact with each other.
3. The adhesive film of claim 1 wherein a third layer is interposed between the primary agent layer and the catalyst layer.
4. The adhesive film of claim 3 wherein said third layer comprises at least member selected from the group consisting of polyvinyl butryal, polysulfone, nylon, and epoxy/amine.
5. The adhesive film of claim 4 wherein said third layer has a thickness of from about 0.1 micrometer to about 50 micrometers.
6. The adhesive film of claim 1 further comprising a release layer.

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-22-

7. The adhesive film of claim 1 wherein the cyanate ester resin is selected from the group consisting of oligomers of bisphenol-A dicyanates, oligomers of tetra o-methyl bisphenol F dicyanates, and oligomers of
5 thiodiphenol dicyanates, said oligomer having a molecular weight of from about 250 to about 1200.

8. The adhesive film of claim 7 wherein the thermoplastic resin is selected from the group consisting
10 of polyvinylformyls, polyvinylacetals, polyvinylbutylols, polyamide, polyimides, polysulfones, polyesters, polyesterimides, polycarbonates, polycarbonatesiloxanes, polyethers, polyallylates, polyimidesiloxanes, phenoxy resins, carboxylated SEBS, acrylic reins, and
15 polyurethanes.

9. The adhesive film of claim 1 wherein the thermoplastic resin used in said primary agent layer is the same as the thermoplastic resin used in said catalyst
20 layer.

10. The adhesive film of claim 1 wherein the primary agent layer has a thickness of from about 1 micrometer to about 100 micrometers, and the catalyst layer has a
25 thickness of from about 0.1 micrometer to about 50 micrometers.



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