Appeal decision

Appeal No. 2019-7148

Appellant	Nissan Chemical Corporation
Patent Attorney	EIMEI INTERNATIONAL PATENT OFFICE

The case of appeal against the examiner's decision of refusal of Japanese Patent Application No. 2016-523538, entitled "Thin-film planarization method, planarized thin-film formation method, and a thin-film formation varnish to be used in these methods" (International publication, December 3, 2015, WO2015/182667) has resulted in the following appeal decision.

Conclusion

The appeal of the case was groundless.

Reason

No. 1 History of the procedures

The international filing date of the Application is May 27, 2015 (claimed priority May 30, 2014), and the subsequent history of the procedures is as follows:

August 16, 2018:	Written statement, and written amendment submitted
September 5, 2018:	Notice of reasons for refusal issued
November 6, 2018:	Written opinion, and written amendment submitted
March 4, 2019:	Examiner's decision of refusal (hereinafter, referred to as "Examiner's decision")
May 31, 2019:	Written demand for trial submitted
June 21, 2019:	Notice of reasons for refusal issued
September 20, 2019:	Written opinion, and written amendment submitted

No. 2 The Invention

The invention accordingly to Claim 1 of the Application (hereinafter, referred to as "the Invention") as specified by the matter disclosed by Claim 1 of the scope of claims amended by the written amendment submitted on September 20, 2019 is as follows:

"[Claim 1]

A varnish for forming charge-transporting thin films comprising an organic compound that is a charge-transporting substance whose molecular mass is 200 to 5,000 and organic solvents wherein the varnish has an flow activation energy of 28 kJ/mol or less (excluding charge-transporting varnishes comprising a charge-transporting substance comprising a charge-transporting monomer or a charge-transporting oligomer or polymer having a number-average molecular weight from 200 to 5,000, or a charge-transporting material comprising the charge transportation substance and a dopant substance, and a mixed sorbent including at least one type of good solvent and at least one type of poor solvent, the absolute value of the boiling point difference ΔT °C of the good solvent and the poor solvent satisfying the relation $|\Delta T| < 20$°C, the viscosity at 25°C being 7.5 mPa•s or less, the surface tension at 23°C being 30.0 to 40.0 mN/m, and the charge-transporting material being dissolved or uniformly dispersed in the mixed solvent)."

No. 3 Outline of notice of reasons for refusal of the body

The reasons for refusal notified by the body with the notice of reasons for refusal dated June 21, 2019 are that [A] the description in the scope of claims of the Application does not comply with the requirements set forth in Article 36(6)(i) of the Patent Act, that [B] since the inventions according to Claims 1 to 17 of the Application are inventions available that were to the public before the priority date (hereinafter, referred to as "the Priority Date") in Japan and abroad through electric telecommunication lines, they fall under Article 29(1)(iii) of the Patent Act and are not patentable, that [C] since the inventions according to Claims 1 to 17 of the Application are inventions that could have been easily made by a person ordinarily skilled in the art of the invention (hereinafter, referred to as "a person skilled in the art") before the filing according to an invention available to the public through electric telecommunication lines before the priority date in Japan and abroad, they are not patentable under the provisions of Article 29(2) of the Patent Act, and that [D] the description in the scope of claims of the Application does not comply with the requirement set forth in Article 36(6)(ii) of the Patent Act.

No. 4 Description in the detailed description of the invention of the Application

The detailed description of the invention of the Application has the following description.

1 "The problem to be solved by the invention

[0008] The Invention has been made based on such situation, and its purposes are to provide a thin-film planarization method, a planarized thin-film formation method, and a varnish that can provide such thin films.

... (Omitted) ...

Description of the embodiment of the invention

[0013] The thin film planarization method of the Invention is characterized by the flow activation energy of the varnish being made 28 kJ/mol or less in forming thin films using a thin-film formation varnish comprising an organic compound and organic solvents.

[0014] The flow activation energy is represented by E (kJ/mol) in Andrade's equation,

$\eta = A\exp (E/RT)$.

Andrade's equation is a formula that represents the relationship between viscosity and temperature, and the lower the flow activation energy, the smaller the temperature dependence of viscosity."

2 "Examples

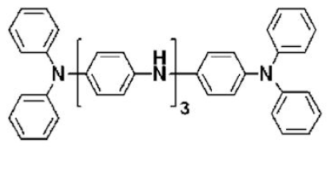
... (Omitted) ...

[0085] [1] Synthesis of components of the charge-transporting varnishes

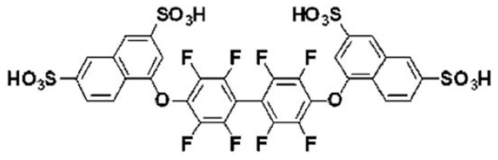
[Synthesis example 1]

The synthesis was carried out using an aniline derivative represented by the following formula (2) and allyl sulfonic acid represented by the following formula (3) in accordance with the methods disclosed in International Publications No. 2013/084664 and No. 2006/025342.

[Chemical formula 5]



(2)



(3)

[0086] [2] Preparation of a charge-transporting varnish

[Example 1] Preparation of charge-transporting varnish A

Charge-transporting varnish A was prepared by dissolving an aniline derivative represented by formula (2) (0.137g) and allyl sulfonic acid represented by formula (3) (0.271g) in 1,3-dimethyl-2-imidazolidinone (6.0g) in a nitrogen atmosphere;

sequentially adding to the obtained solution diethylene glycol (6.0g) and diethylene glycol monomethyl ether (8.0g), and agitating the solution.

[0087] [Example 2] Preparation of charge-transporting varnish B

Charge-transporting varnish B was prepared with the same method as in Example 1 except that the solvent was changed to 1,3-dimethyl-2-imidazolidinone (6.0g), dipropylene glycol monomethyl ether (9.0g), and hexylene glycol (5.0 g).

[0088] [Example 3] Preparation of charge-transporting varnish C

Charge-transporting varnish C was prepared with the same method as in Example 1 except that the solvent was changed to 1,3-dimethyl-2-imidazolidinone (6.0g), diethylene glycol (10.0g), and diethylene glycol monomethyl ether (4.0 g).

[0089] [Example 4] Preparation of charge-transporting varnish D

Charge-transporting varnish D was prepared with the same method as in Example 1 except that the solvent was changed to 1,3-dimethyl-2-imidazolidinone (8.0g), 2,3-butanediol (9.0g), and diethylene glycol monoethyl ether acetate (3.0 g).

[0090] [Example 5] Preparation of charge-transporting varnish E

Charge-transporting varnish E was prepared with the same method as in Example 1 except that the solvent was changed to 1,3-dimethyl-2-imidazolidinone (6.6g), 2,3-butanediol (8.0g), and dipropylene glycol monomethyl ether (5.4 g).

... (Omitted) ...

[0092] [3] Calculation of the flow activation energy

Using a rheometer, the viscosity (η) of each of charge-transporting varnishes A to F at 20 to 100°C was measured. Since Andrade's equation can be transformed as shown below, the flow activation energy E of each varnish was calculated by plotting with $I\ n\eta$ to $1/T$ using obtained measured values. For the calculation, the least squares method was used.

$I\ n\eta = I\ nA + (E/RT)$

In addition, with the same method, the viscosity (η) of only each of the solvents used for preparation of charge-transporting varnishes A to F was measured and the flow activation energy Es of each solvents was calculated. The results are shown in Table 1.

[0093] [4] Evaluation of flatness in a pixel

Film formation was carried out by applying charge-transporting varnishes A to F to ITO substrate with structures with 50 x 100 μm pixel width (between structures) using a positive type sensitive polyimide, and the films were dried by heating to 80°C on a hotplate for one minute in the air, and heat-burning was carried out at 230°C for 15 minutes. Maximum step (Rmax) of the film in the pixel portion was measured with a surface roughness measuring instrument, Surfcoder ET-4000. The results are

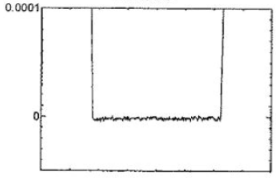
summarized in Table 1.　A film profile in the pixel is shown in Figure 1.
[0094] [Table 1]

	実施例 1	実施例 2	実施例 3	実施例 4	実施例 5	比較例 1
	ワニスA	ワニスB	ワニスC	ワニスD	ワニスE	ワニスF
E（kJ/mol）	22.1	23.0	24.6	25.3	27.8	29.3
E_s (kJ/mol)	21.7	21.9	23.6	25.6	27.0	28.6
R_{max}（nm）	1.4	5.4	7.1	10.9	14.0	22.7

実施例	Example
比較例	Comparative Example
ワニス	Varnish

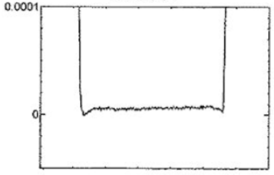
[0095] As indicated in Table 1, since the film prepared with charge-transporting varnish A having the smallest flow flow activation energy had the smallest maximum step, and the larger the flow flow activation energy, the larger the maximum step, it was found that the flatness of a film can be improved by making the activity energy of charge-transporting varnish lower."

3. "[Figure 1]



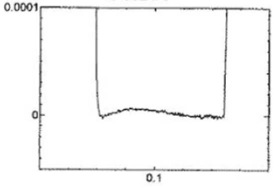
実施例1

Rmax: 1.4 nm



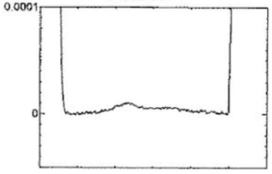
実施例2

Rmax: 5.4 nm



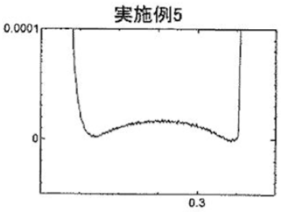
実施例3

Rmax: 7.1 nm



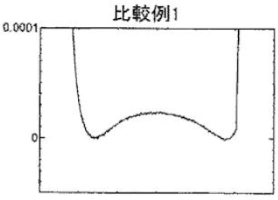
実施例4

Rmax: 10.9 nm



実施例5

Rmax: 14.0 nm



比較例1

Rmax: 22.7 nm

"

実施例　Example
比較例　Comparative Example

No.5 Description in Cited Document

It has been acknowledged that International Publication No. 2013/129249 (hereinafter, referred to as "Cited Document") that became available to the public on September 6, 2013 before the filing of the Application in Japan and abroad through electric telecommunication lines is a document that discloses the technical level before the filing of the Application, and the Cited Document has the following described matter.

"Description of the embodiment of the invention

[0010] The present invention is further explained in detail below.

The charge-transporting varnish of the Invention comprises a charge-transporting substance comprising a charge-transporting monomer or a charge-transporting oligomer or polymer having a number-average molecular weight from 200 to 500,000, or a charge-transporting material comprising the charge transportation substance and a dopant substance, and a mixed sorbent including at least one type of good solvent and at least one type of poor solvent, the absolute value of the boiling point difference ΔT °C of the good solvent and the poor solvent satisfying the relation $|\Delta T| < 20°C$, the viscosity at 25°C being 7.5 mPa•s or less, the surface tension at 23°C being 30.0 to 40.0 mN/m, and the charge-transporting material being dissolved or uniformly dispersed in the mixed solvent.

... (Omitted) ...

[0048] In the charge-transporting varnish of the invention, the amounts of the used good solvent and poor solvent cannot be simply defined because they vary depending on the types of used solvents as well as the types, used amounts, etc. of the charge-transporting substance, etc. so that the viscosity and the surface tension of the varnish satisfy the above range, but, generally, the amount of the used good solvent is 1 to 90% by mass (poor solvent 10 to 99% by mass) to the total amount of the solvent, preferably 10 to 90% by mass (poor solvent 10 to 90% by mass), more preferably 20 to 90% by mass (poor solvent 10 to 80% by mass), and still more preferably 30 to 90% by mass (poor solvent 10 to 70% by mass).

... (Omitted) ...

[0096] Varnishes prepared in each Example and Comparative Example were applied to an ITO solid substrate (longitudinally 120 mm x 120 mm, thickness 0.7 mm) using a slit die coater, dried in the air on a 50°C hot plate for 5 minutes, and burnt on a 230°C hot plate for 15 minutes to form thin films on the substrate.

The ITO substrate was used after removing impurities on the surface using O2 plasma cleaning equipment (150 W, 30 seconds), and coating conditions of the slit coater were a coating area of 120 mm x 120 mm, gap 20 µm, coating waiting time 4 sec, coating speed 10 mm/sec, and target film thickness 30 nm.

[0097] Next, the film thickness distribution of the obtained thin films was evaluated. Evaluation was carried out by measuring the film thickness at five points in total, four corners and the point around the center of thin films that had been made square, and quantifying the in-plane uniformity of the film thickness at the five points as the film thickness distribution. Tables 1 to 4 show the viscosity and the surface tension of each varnish, the boiling point difference [boiling point of the good solvent (°C) − boiling point of the poor solvent (°C)] of good solvent and poor solvent used for preparing varnishes, and their absolute values, as well as result of evaluation of the film thickness distribution.

Examples in which the impact of the viscosity of varnish on the film thickness distribution can be easily evaluated were extracted and enumerated in Table 1, examples in which the impact of the surface tension of varnish on the film thickness distribution can easily be evaluated were extracted and enumerated in Table 2, and examples in which the impact of the boiling point difference between good solvent and poor solvent on the film thickness distribution can easily be evaluated were extracted and enumerated in Table 3.

Table 4 indicates the result of Comparative Examples 11 and 12 for a single solvent composition.

[0098] [Table 1]

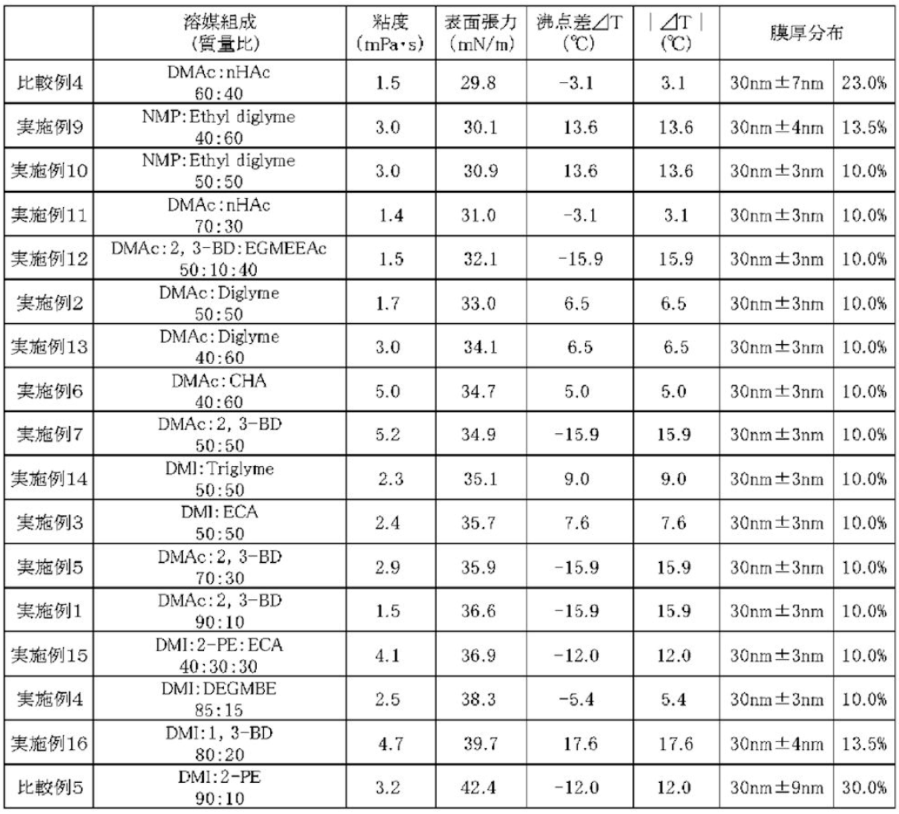
	溶媒組成 (質量比)	粘度 (mPa･s)	表面張力 (mN/m)	沸点差⊿T (℃)	|⊿T| (℃)	膜厚分布	
比較例3	DMAc:2, 3-BD 30:70	12.7	34.3	-15.9	15.9	30nm±11nm	36.5%
比較例2	DMAc:2, 3-BD 35:65	10.8	34.7	-15.9	15.9	30nm±8nm	26.6%
比較例1	DMAc:2, 3-BD 40:60	8.1	34.9	-15.9	15.9	30nm±5nm	16.7%
実施例8	DMAc:2, 3-BD 45:55	6.8	35.3	-15.9	15.9	30nm±3nm	10.0%
実施例7	DMAc:2, 3-BD 50:50	5.2	34.9	-15.9	15.9	30nm±3nm	10.0%
実施例6	DMAc:CHA 40:60	5.0	34.7	5.0	5.0	30nm±3nm	10.0%
実施例5	DMAc:2, 3-BD 70:30	2.9	35.9	-15.9	15.9	30nm±3nm	10.0%
実施例4	DMI:DEGMBE 85:15	2.5	38.3	-5.4	5.4	30nm±3nm	10.0%
実施例3	DMI:ECA 50:50	2.4	35.7	7.6	7.6	30nm±3nm	10.0%
実施例2	DMAc:Diglyme 50:50	1.7	33.0	6.5	6.5	30nm±3nm	10.0%
実施例1	DMAc:2, 3-BD 90:10	1.5	36.6	-15.9	15.9	30nm±3nm	10.0%

溶媒組成	Solvent composition
（質量比）	(Mass ratio)
粘度	Viscosity
表面張力	Surface tension
沸点差	Boiling point difference
膜厚分布	Film thickness distribution
比較例	Comparative example
実施例	Example

[0099] As indicated in Table 1, it can be understood that thin films made with varnishes prepared in Examples 1 to 8 having a viscosity of 7.5 mPa•s or less have a smaller variation in film thickness as compared with thin films made with varnishes prepared in

Comparative Examples 1 to 3 whose viscosity is higher than 7.5 mPa•s.

[0100] [Table 2]



	溶媒組成 (質量比)	粘度 (mPa·s)	表面張力 (mN/m)	沸点差⊿T (℃)	|⊿T| (℃)	膜厚分布	
比較例4	DMAc:nHAc 60:40	1.5	29.8	−3.1	3.1	30nm±7nm	23.0%
実施例9	NMP:Ethyl diglyme 40:60	3.0	30.1	13.6	13.6	30nm±4nm	13.5%
実施例10	NMP:Ethyl diglyme 50:50	3.0	30.9	13.6	13.6	30nm±3nm	10.0%
実施例11	DMAc:nHAc 70:30	1.4	31.0	−3.1	3.1	30nm±3nm	10.0%
実施例12	DMAc:2, 3-BD:EGMEEAc 50:10:40	1.5	32.1	−15.9	15.9	30nm±3nm	10.0%
実施例2	DMAc:Diglyme 50:50	1.7	33.0	6.5	6.5	30nm±3nm	10.0%
実施例13	DMAc:Diglyme 40:60	3.0	34.1	6.5	6.5	30nm±3nm	10.0%
実施例6	DMAc:CHA 40:60	5.0	34.7	5.0	5.0	30nm±3nm	10.0%
実施例7	DMAc:2, 3-BD 50:50	5.2	34.9	−15.9	15.9	30nm±3nm	10.0%
実施例14	DMI:Triglyme 50:50	2.3	35.1	9.0	9.0	30nm±3nm	10.0%
実施例3	DMI:ECA 50:50	2.4	35.7	7.6	7.6	30nm±3nm	10.0%
実施例5	DMAc:2, 3-BD 70:30	2.9	35.9	−15.9	15.9	30nm±3nm	10.0%
実施例1	DMAc:2, 3-BD 90:10	1.5	36.6	−15.9	15.9	30nm±3nm	10.0%
実施例15	DMI:2-PE:ECA 40:30:30	4.1	36.9	−12.0	12.0	30nm±3nm	10.0%
実施例4	DMI:DEGMBE 85:15	2.5	38.3	−5.4	5.4	30nm±3nm	10.0%
実施例16	DMI:1, 3-BD 80:20	4.7	39.7	17.6	17.6	30nm±4nm	13.5%
比較例5	DMI:2-PE 90:10	3.2	42.4	−12.0	12.0	30nm±9nm	30.0%

溶媒組成	Solvent composition
（質量比）	(Mass ratio)
粘度	Viscosity
表面張力	Surface tension
沸点差	Boiling point difference
膜厚分布	Film thickness distribution
比較例	Comparative example
実施例	Example

[0101] As indicated in Table 2, it can be understood that thin films made with varnishes prepared in Examples in which the surface tension of varnishes is within the range of 30.0 to 40.0 mN/m have a smaller variation in film thickness as compared with thin films made with varnishes prepared in Comparative Examples 4 and 5 in which the surface tension of varnishes is outside the above range.

[0102] [Table 3]

	溶媒組成（質量比）	粘度（mPa･s）	表面張力（mN/m）	沸点差⊿T（℃）	｜⊿T｜（℃）	膜厚分布	
比較例6	DMAc:TEGMME 70:30	1.8	37.5	-82.9	82.9	30nm±7nm	23.0%
比較例7	DMI:TEGMEE 50:50	5.0	36.5	-30.9	30.9	30nm±6nm	20.0%
比較例8	DMI:DEGMBEAc 60:40	3.1	34.4	-21.8	21.8	30nm±5nm	16.7%
実施例17	DMI:TPGMME 60:40	6.2	30.0	-18.0	18.0	30nm±3nm	10.0%
実施例7	DMAc:2, 3-BD 50:50	5.2	34.9	-15.9	15.9	30nm±3nm	10.0%
実施例5	DMAc:2, 3-BD 70:30	2.9	35.9	-15.9	15.9	30nm±3nm	10.0%
実施例1	DMAc:2, 3-BD 90:10	1.5	36.6	-15.9	15.9	30nm±3nm	10.0%
実施例4	DMI:DEGMBE 85:15	2.5	38.3	-5.4	5.4	30nm±3nm	10.0%
実施例2	DMAc:Diglyme 50:50	1.7	33.0	6.5	6.5	30nm±3nm	10.0%
実施例6	DMAc:CHA 40:60	5.0	34.7	5.0	5.0	30nm±3nm	10.0%
実施例3	DMI:ECA 50:50	2.4	35.7	7.6	7.6	30nm±3nm	10.0%
実施例18	DMAc:EGMEEAc 70:30	0.9	33.5	9.3	9.3	30nm±3nm	10.0%
実施例19	DMI:EGMHE 60:40	3.4	32.4	16.9	16.9	30nm±3nm	10.0%
比較例9	DMI:DEGMEE 50:50	4.1	36.3	30.0	30.0	30nm±6nm	20.0%
比較例10	DMI:PGME 80:20	2.4	37.5	105.0	105.0	30nm±8nm	26.7%

溶媒組成	Solvent composition
（質量比）	(Mass ratio)
粘度	Viscosity
表面張力	Surface tension
沸点差	Boiling point difference
膜厚分布	Film thickness distribution
比較例	Comparative example
実施例	Example

[0103] As indicated in Table 3, it can be understood that thin films made with varnishes prepared in Examples in which the absolute value of the boiling point difference between the good solvent and the poor solvent is |ΔT| < 20°C have a smaller variation in film thickness as compared with thin films made with varnishes prepared in Comparative Examples 6 to 10 in which |ΔT| is 20°C or higher."

No. 6 Judgment

1 According to the description in paragraph [0008] of the specification of the Invention, it is recognized that the problem to be solved by the Invention is "to provide thin films with excellent flatness." On the other hand, in paragraph [0013] in the detailed description of the invention, there is a description that, as a method to solve the above problem, "flow activation energy" of the "varnish for forming charge-transporting thin films" was made "28 kJ/mol or less."

Although it is stated in paragraph [0014] in the specification of the Invention that "the lower the flow activation energy, the smaller the temperature dependence of the viscosity," the relationship between the flow activation energy of a varnish and the flatness of the thin films formed from the varnish is not mentioned and the relationship is not clear in the light of common technical knowledge as of the filing of the application.

For example, as stated in paragraph [0048] of the Cited Documents that "the amounts of used good solvent and poor solvent cannot be simply defined because they vary depending on the types of used solvents as well as the types, used amounts, etc. of the charge-transporting substance, etc. so that the viscosity and the surface tension of the varnish satisfy the above range," and, in paragraph [0097], that "the viscosity of varnish influences the film thickness distribution," that "the surface tension of varnish influences the film thickness distribution" and that "the boiling point difference between the good solvent and the poor solvent influences the film thickness distribution," it is recognized that the flatness of a thin film is not simply determined only by the flow activation energy, and it depends on the viscosity and the surface tension of varnish, as well as the boiling point of solvent and material composition that affects such indexes. Then, it is considered that the preferred upper limit value of the flow activation energy of a varnish varies depending on the material composition.

Only varnishes for forming charge-transporting thin films described in Examples 1 to 5 are mentioned in the specification of the Invention as "varnishes for forming charge-transporting thin films" whose "flow activation energy is 28 kJ/mol or less," and it cannot be deemed that it has concretely confirmed that, even with varnishes for forming charge-transporting thin films consisting of materials other than those mentioned in Examples 1 to 5, the above problem can be solved by making the "flow activation energy" "28 kJ/mol or less" (note by the body: For example, since the "organic compound" in each Example is limited to "an aniline derivative represented by the following formula (2)" whose molecular weight is around 685 as described in paragraph [0085] in the detailed description of the invention of the Application, it cannot be deemed that it has been concretely confirmed that the above problem can be solved even in the case of an "organic compound" whose molecular weight is 5000 or in the case of an "organic compound" that has a completely different structure.).

Then, even if the description in the specification of the Invention is considered generally, no ground to judge that any "varnish for forming charge-transporting thin films" whose material composition has not been specified can solve the problem if the varnish satisfies the conditions defined in Claim 1. In addition, it also cannot be deemed that a person skilled in the art can recognize in the light of common technical knowledge as of the filing of the application that the problem to be solved by the invention can be solved.

Therefore, the Invention cannot be deemed to be within the scope in which a person skilled in the art can recognize that the problem to be solved by the invention can be solved with the description in the detailed description of the invention, and the Invention cannot be deemed to be within the scope in which a person skilled in the art can recognize that the problem to be solved by the invention can be solved in the light of common technical knowledge as of the filing even without the description or suggestion.

2 In this regard, Applicant asserts in its written opinion submitted on September 20, 2019 that "the upper limit of the molecular weight of the charge-transporting substance has been specified with the above amendment. From matters disclosed in Examples in the Application, the person skilled in the art can easily understand that the effect of the Invention can be obtained if the charge-transporting substance of which molecular weight is in this range is used."

As stated in above item 1, however, since it is recognized that the preferred upper limit value of the flow activation energy of a varnish varies depending on its material composition, no ground to judge that the problem to be solved by the Invention can be solved can be found just by specifying the upper limit of the molecular weight of the charge-transporting substance as described in Claim 1.

Therefore, Applicant's opinion cannot be accepted.

No. 7 Closing

As described above, in this application, the descriptions in the scope of claims do not comply with the requirements set forth in Article 36(6)(i) of the Patent Act.

Therefore, the appeal decision shall be made as described in the conclusion.

November 19, 2019

Chief administrative judge:	HIGUCHI, Nobuhiro
Administrative judge:	TAKAMATSU, Dai
Administrative judge:	MIYAZAWA, Hiroshi