

## Appeal decision

Appeal No. 2015-5639

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The case of Appeals against an Examiner's Decision of refusal of Japanese Patent Application No. 2011-537879 "Optical brightening agent compositions for high quality ink jet printing" [International Publication on June 3, 2010, WO 2010-060570, and National Publication of International Patent Application No. 2012-509796 published on April 26, 2012] has resulted in the following trial decision.

#### Conclusion

The demand for trial of the case was groundless.

#### Reason

##### No. 1 Outline of case

##### 1 History of the procedures

The present application is an application that was filed on November 20, 2009 (Heisei 21) (claim of the priority on November 27, 2008 (Heisei 20) in the European Patent Office, and on July 2, 2009 (Heisei 21) in the European Patent Office) as an international filing date, and the outline of the history of the subsequent procedures is as follows:

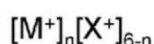
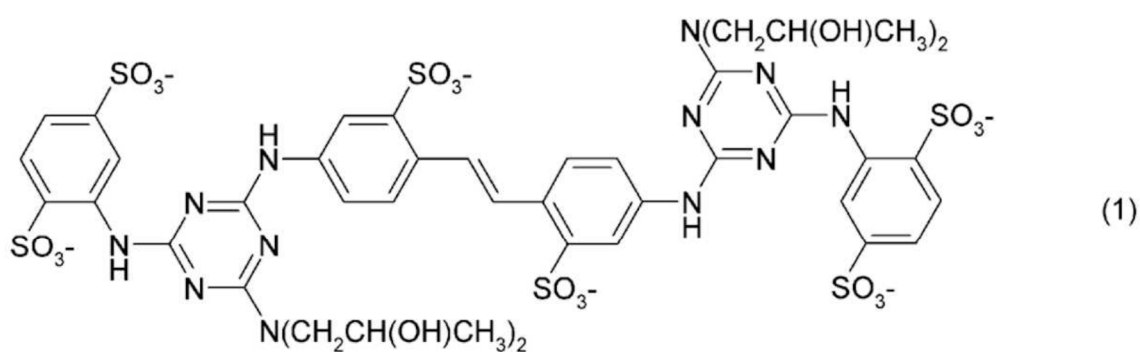
November 19, 2012:	Written amendment
December 26, 2013:	Notice of reasons for refusal (dispatched on January 28, 2014)
May 27, 2014:	Written opinion
November 21, 2014:	Decision of refusal (delivered on November 26, 2014)
March 25, 2015:	Demand for trial

##### 2 The Invention

The invention (hereinafter referred to as the "Invention") described in Claim 1 in claims of the present application is as follows:

"A sizing composition that optically brightens a substrate for ink jet printing, comprising:

- (a) at least one binder;
  - (b) at least one divalent metal salt, the at least one divalent metal salt being selected from the group consisting of calcium chloride, magnesium chloride, calcium bromide, magnesium bromide, calcium iodide, magnesium iodide, calcium nitrate, magnesium nitrate, calcium formate, magnesium formate, calcium acetate, magnesium acetate, calcium sulphate, magnesium sulphate, calcium thiosulphate and magnesium thiosulphate, and mixtures of said compounds,
  - (c) water; and
  - (d) at least one optical brightener of formula (1).
- [Formula 1]



(In the formula,

M and X are identical or different and independently from each other selected from the group consisting of hydrogen, an alkali metal cation, ammonium, and ammonium which is mono-, di- or trisubstituted by a C<sub>1</sub>-C<sub>4</sub> linear or branched alkyl radical, ammonium which is mono-, di- or trisubstituted by a C<sub>1</sub>-C<sub>4</sub> linear or branched hydroxyalkyl radical, and mixtures of said compounds, and n is in the range from 0 to 6.)"

### 3 Reasons for refusal stated in the examiner's decision

The reason for refusal stated in the examiner's decision is briefly that the Invention could have been easily made by a person who had the ordinary skill in the art belonging to the Invention prior to the filing of the application, based on the invention described in Cited Document 1 or Cited Document 2 in Japan or a foreign country before the date of the claim of the priority (hereinafter referred to as "priority date"), and on the well-known art, and accordingly the patent should not be granted under the provisions of Article 29 (2) of the Patent Act.

Cited Document 1: Japanese Unexamined Patent Application Publication No. 2003-266923

Cited Document 2: Japanese Unexamined Patent Application Publication No. 2002-348494

### No. 2 Judgment of collegial body

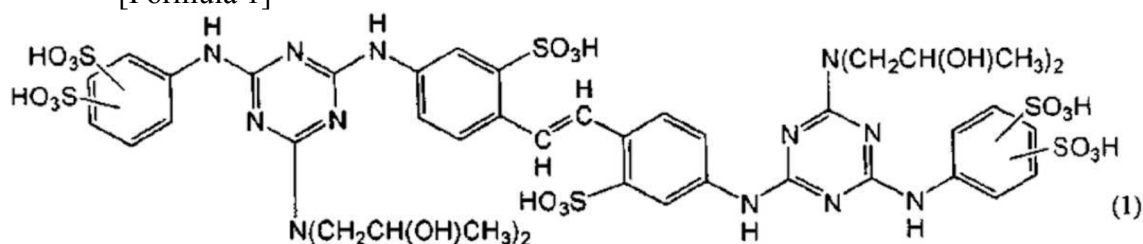
#### 1 Cited documents and cited invention

##### (1) Description of Cited Document 1

The following matters are described in Cited Document 1. Underlines are provided by this collegial body (Hereinafter the same.).

A. "[Claim 1] Ink jet archival paper having a fluorescent brightener represented by the following formula (1) in a form of a free acid.

[Formula 1]



(In the formula, a cation corresponding to the sulfonic group is any of cations of an alkali metal or alkaline earth metal, or an ammonium ion)  
[Claim 2] The ink jet archival paper according to claim 1, having a cationic ink fixing agent."

B. "[0001]

[Field of the Invention] The present invention relates to ink jet archival paper, and more specifically, to a method for producing the ink jet archival paper that exhibits excellent ink absorbency and ink fixability, provides an image quality of high quality, and has high whiteness.

[0002]

[Prior art] A printer employing an ink jet system facilitates multiple coloring, shows clearness of color, produces quiet sound at the time of operation, and provides a recorded matter of high quality; and accordingly has become popular in recent years. Various types of dyes are used in the ink of the ink jet, including an ink in which an oil-soluble dye is adjusted by a solution containing an organic solvent, an ink in which a water-soluble dye is adjusted by water, an ink in which a pigment dye is dispersed, and the like. Among the inks, the ink using the water-soluble dye is a mainstream for the ink jet printer, by virtue of being capable of printing a recorded matter that has clearness of printing and high quality of a picture tone, at quick speed. An anionic dye is used in the ink of the ink jet printer, which uses the water-soluble dye. Because of this, in archival paper of high quality for an ink jet printer, in order to suppress the bleeding of ink on the paper, a cationic ink fixing agent, water resistant additive, or the like are used in many cases when the ink jet archival paper is produced. In addition, the archival paper is required to have clear color developability for the image, and accordingly is also required to have whiteness of high quality. Because of this, there are produced sheets of ink jet archival paper which are dyed into high whiteness with the use of various fluorescent brighteners. As for the fluorescent brightener that is used at this time, in many cases an anionic water-soluble fluorescent brightener is used, from the viewpoint of characteristics in a production process. However, when the above-described cationic fixing agent and the anionic fluorescent brightener are used at the same time, there have been significant problems that the ink fixing agent and the fluorescent brightener are combined in the solution to thereby become insoluble in water, resulting in precipitation of crystal, and that there occurs such a phenomenon that high whiteness required in dyeing is not obtained.

[0003]

[Problem to be solved by the invention] An object of the present invention is to solve the above-described problems, and provide a method for producing receptive paper for the ink jet, which has whiteness of high quality, by simultaneously using a cationic ink fixing agent and an anionic water-soluble fluorescent brightener."

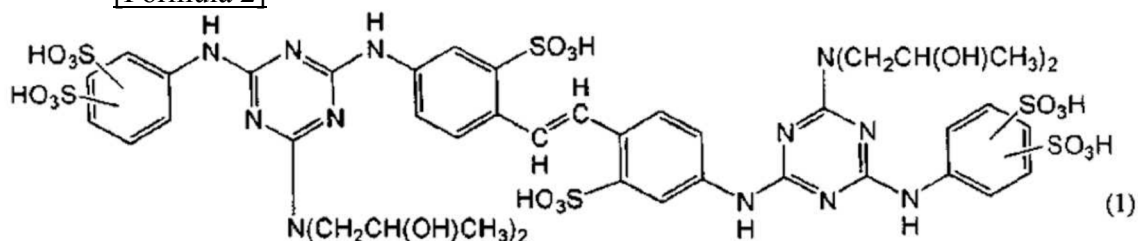
C. "[0004]

[Means for solving problem] The present inventors have found that the above-described problems are solved by using a fluorescent brightener having a specific structure, when archival paper for the ink jet is dyed, and thereby have completed the present invention. Specifically, the present invention relates to: (1) ink jet archival paper having a fluorescent brightener expressed by the following formula (1) in a form

of a free acid,

[0005]

[Formula 2]



[0006] (In the formula, a cation corresponding to the sulfonic group is any of cations of an alkali metal and an alkaline earth metal, or an ammonium ion);

(2) the ink jet archival paper described in (1), which has a cationic ink fixing agent; and  
(3) the ink jet archival paper described in (1) or (2), onto which a coating liquid for an ink-receptive layer is applied."

D. "[0007]

[Mode for carrying out the invention] The present invention will be described in detail below. As for the fluorescent brightener expressed by formula (1), the structure thereof has already been disclosed in Belgium patent No. 719065, and is obtained, for instance, by steps of making 1 mole of 4,4'-diaminostilbene-2,2'-disulfonic acid react with 2 moles of cyanuric chloride, making the obtained compound react with 2 moles of aniline disulfonic acid, and making the obtained compound react with 2 moles of di-2-propanolamine. Alternatively, the fluorescent brightener is obtained by steps of making a compound which has been obtained by a step of making 2 moles of aniline disulfonic acid react with 2 moles of cyanuric chloride react with 1 mole of 4,4'-diaminostilbene-2,2'-disulfonic acid, and making the obtained compound react with 2 moles of di-2-propanolamine.

[0008] A reaction liquid of the fluorescent brightener expressed by formula (1), which has been obtained by those methods, can be acid precipitated by hydrochloric acid, sulfuric acid, nitric acid, or the like, and taken out as a crystal of a free acid; or can be salted out by an inorganic salt such as sodium chloride, potassium chloride, calcium chloride, or sodium sulfate, and thereby taken out as a crystal of an alkali metal salt or an alkaline earth metal salt such as a sodium salt, a potassium salt, or a calcium salt.

[0009] The obtained free acid of the fluorescent brightener expressed by formula (1) is dissolved in water together with a desired amount of sodium hydroxide; or an alkali metal salt, an alkaline earth metal salt, an ammonium salt, or the like of the fluorescent brightener expressed by formula (1) is dissolved in water; and thereby an aqueous solution of usually 10-40 mass% of the fluorescent brightener is obtained, which is used in the present invention. Alternatively, the fluorescent brightener to be used in the present invention is obtained also by using the obtained reaction liquid as is or after having been concentrated.

[0010] As for a substrate of the ink jet archival paper that is used in the present invention, there is applied a sheet of paper which is formed from a pulp raw material that mainly contains a cellulose fiber, and no particular limitations are imposed on the composition and the production process. For instance, there is used a sheet of paper which is produced by a wet papermaking process. Specifically, a wood pulp such as kraft pulp, sulphite pulp, or semichemical pulp is used as a pulp raw material, which is

produced by using a needle-leaved tree or a broad-leaved tree independently or in appropriate combination for blending. Bleached pulp is preferably used, in order that a clear image is printed on the receptive paper. In addition, it is also possible to use a recycled pulp or a nonwood pulp such as bagasse, kenaf, cotton, hemp, esparto, bamboo, or straw.

[0011] It is possible to use an internal paper strengthening agent such as starch, modified starch, carboxymethyl cellulose, polyacrylamide, or a styrene-based resin; a coloring agent; or a fixing agent such as aluminum sulfate or polyacrylamide, as a chemical to be added to the pulp raw material; and further to use dialdehyde starch, a melamine resin, an antistatic agent, a water repellent agent, a lubricant, a sizing agent, a pH adjuster, an antiseptic agent, a dispersing agent, a slip additive, a surface active agent, or the like as a coating chemical, as needed, in addition to the above-described surface paper strengthening agent.

[0012] The fluorescent brightener expressed by formula (1) of the present invention and an aqueous solution thereof are suitable for dyeing of a cellulose-based material such as paper or pulp, and is particularly suitable for dyeing of ink jet archival paper. As for the ink jet archival paper, an overcoat layer such as an ink-receptive layer or a gloss layer is provided, as needed, after beating of the pulp, papermaking, and size pressing.

[0013] An internal addition method is a method of adding a fluorescent brightener in a step after the beating of the pulp and before the papermaking, and thereby is capable of coloring the beaten pulp. The method includes the steps of: firstly beating pulp to a predetermined degree of beating by a pulper, a refiner, or the like to form a pulp slurry; adding the fluorescent brightener expressed by formula (1) which usually corresponds to 0.01 to 4.0% (net content of brightener per mass of dried paper) usually at 10 to 40°C, or an aqueous solution thereof; further adding a normal sizing agent, aluminum sulfate, a paper strengthening agent, a sticking agent, and the like, as needed; and making paper and drying the paper by an ordinary method. Then, there is obtained colored paper which has been fluorescent-whitened.

[0014] The size press by an external addition method is a method of adding a fluorescent brightener to a size pressing liquid in a size pressing step after papermaking. The method includes the steps of: after having made paper, applying to the paper a size press coating liquid that contains the fluorescent brightener and the ink fixing agent, in a step of drying the paper with a cylinder drier, by a size pressing machine that is arranged in a middle portion at which a large number of cylinder driers are arranged, and after that, drying the coating liquid. Thereby, there is obtained the paper which has been fluorescent-whitened. The above-described size press coating liquid is prepared by a process of appropriately mixing a fluorescent brightener, an ink fixing agent, the fluorescent brightener expressed by formula (1) or the aqueous solution thereof, starch, PVA, CMC, a surface sizing agent, water, and the like; and the content of the fluorescent brightener expressed by formula (1) in the size press coating liquid is usually 0.001 to 2.0% (net content); and an amount of the size press coating liquid to be coated is usually 0.1 to 3 g/m<sup>2</sup> (dry mass) of dried paper."

E. "[0018] Usable cationic ink fixing agents include: a higher fatty amine, a quaternary-ammonium-salt type compound, an ethylene oxide addition product of a secondary alkylamine, a cationic polymer compound, and inorganic particles whose

surfaces are cationically charged."

F. "[0020] The inorganic particles whose surfaces are cationically charged include, for instance, fine particles of alumina, and silica particles that are formed by a reaction of a compound having both of a cationic group and a group capable of reacting with the surface of a silica particle thereon. An amount of the cationic substances to be used in the case where the cationic substances are applied onto paper is approximately 0.1 to 10 g/m<sup>2</sup>, and preferably approximately 0.5 to 5 g/m<sup>2</sup>."

G. "[0021]

The compound expressed by formula (1) in the present invention can produce ink jet archival paper of high whiteness by being used in size press by the internal addition method and the external addition method, in overcoating by the external addition method, and in the ink-receptive layer. Among these uses, in the size press by the external addition method, the overcoating by the external addition method, and the ink-receptive layer, there can also be produced ink jet archival paper that has high ink absorbability and high whiteness, by concomitantly using a cationic ink fixing agent."

H. "[0023]

[Examples] The present invention will be more specifically described below with reference to examples and comparative examples, but the present invention is not limited to these examples.

[0024] Example 1

Twenty parts by mass of cyanuric chloride was dispersed with the use of 80 parts by mass of water, 40 parts by mass of ice, and 0.1 parts by mass of a nonionic dispersing agent. To this dispersion liquid, 32.7 parts by mass of aniline-2,5-disulfonic acid was added. The dispersion liquid was cooled to a temperature of 5°C by an addition of ice, and the temperature was raised from 5.0 to 25°C for 3 hours while the pH was kept at 4.0 to 4.5 by the addition of a sodium hydroxide solution. To this solution, an aqueous solution (130 parts by mass) was added dropwise for 3 hours, in which 19.5 parts by mass of 4,4'-diaminostilbene-2,2'-disulfonic acid was dissolved and the pH was adjusted so as to become 9.5 to 10.5 by sodium hydroxide. In the meantime, the sodium hydroxide solution was added to the solution so that the pH was kept at 4.0 to 4.5, and the temperature was gradually raised from 35 to 50°C. To this solution, 18 parts by mass of di-2-propanolamine were added, and the resultant solution was reacted at 80°C for 3 hours while the pH was kept at 8.5 to 9.0 by the addition of a sodium hydroxide solution. Hydrochloric acid was added to the obtained reaction liquid to adjust the pH to 0.6, crystals that precipitated were filtered, and the compound expressed by formula (1) was obtained. To 50 parts by mass of these crystals that contain water, 80 parts by mass of water was added, the pH was adjusted to 8.5 by sodium hydroxide, and an aqueous solution containing 15 mass% of a sodium salt of the compound expressed by formula (1) was obtained. ( $\lambda_{\text{max}}$  in water = 348 nm)

[0025] (Overcoating by external addition method with the use of coating liquid for receptive layer for ink jet archival paper)

Example 2

Compounds were used which were 100 parts of ultrafine powder of silica (product of MIZUSAWA INDUSTRIAL CHEMICALS, LTD., trade name:

MIZUKASIL P-78A), 30 parts of polyvinyl alcohol (product of Nippon Synthetic Chemical Industry Co., Ltd., trade name: GOHSENL NM-11), 10 parts in terms of a solid content of polydiallyl dimethyl ammonium chloride that is a cationic dye fixing agent (quaternary ammonium salt of polymer) (product of SENKA corporation, trade name: HP-126A), and 0.9 parts of an aqueous solution of a sodium salt of the compound expressed by formula (1), in terms of the net content of the fluorescent brightener; and were adjusted by water so that the coating liquid concentration (solid content) became 13%. Thus, the coating liquid for the receptive layer was obtained. This coating liquid for the ink-receptive layer was applied onto a sheet of commercial neutral PPC paper with a bar coater so that the dry mass became 5 g/m<sup>2</sup>, and was dried at 120°C to produce the fluorescent-whitened paper provided with a receptive layer. The whiteness degree ( $\Delta W$ ) obtained by measuring the color with the use of a spectrum whiteness colorimeter (SC-10W: product of Suga Test Instruments Co., Ltd.) is shown in Table-1."

I. "[0032]

[Effect of the Invention] According to the present invention, ink jet archival paper of excellent printing characteristics is provided, which has adequate ink absorbency as well as whiteness of high quality, and on which a clear image quality free from bleeding of ink or the like is obtained."

(2) Invention described in Cited Document 1

A. Cited Document 1A invention

In paragraph [0024] of Cited Document 1, Example 1 is disclosed as an example of "an aqueous solution containing ... a sodium salt of the compound expressed by formula (1)", and in paragraph [0025], Example 2 is disclosed as an example of the overcoating by the external addition method on the ink jet archival paper, with the use of "an aqueous solution of the sodium salt of the compound expressed by formula (1)".

Then, in Cited Document 1, the following invention (hereinafter referred to as "Cited Document 1A invention") is described. (The paragraph number is written together. Hereinafter the same.).

"[0025] A coating liquid for a receptive layer of ink jet archival paper is a coating liquid for a receptive layer for overcoating by an external addition method on ink jet archival paper, and was obtained by the steps of:

[0024] (A) dispersing 20 parts by mass of cyanuric chloride with the use of 80 parts by mass of water, 40 parts by mass of ice, and 0.1 parts by mass of a nonionic dispersing agent; (B) adding 32.7 parts by mass of aniline-2,5-disulfonic acid to this dispersion liquid; (C) cooling the resultant dispersion liquid to a temperature of 5°C by adding ice, and raising the temperature from 5.0 to 25°C over 3 hours while keeping the pH at 4.0 to 4.5 by adding a sodium hydroxide solution; (D) adding the aqueous solution (130 parts by mass) to this solution dropwise for 3 hours, in which 19.5 parts by mass of 4,4'-diaminostilbene-2,2'-disulfonic acid was dissolved and the pH was adjusted so as to become 9.5 to 10.5 by sodium hydroxide; (E) in the meantime, adding the sodium hydroxide solution to the solution to keep the pH at 4.0 to 4.5, and gradually raising the temperature from 35 to 50°C; (F) adding 18 parts by mass of di-2-propanolamine to this solution, and making the resultant solution react at 80°C for 3 hours while keeping the pH at 8.5 to 9.0 by adding a sodium hydroxide solution; (G)

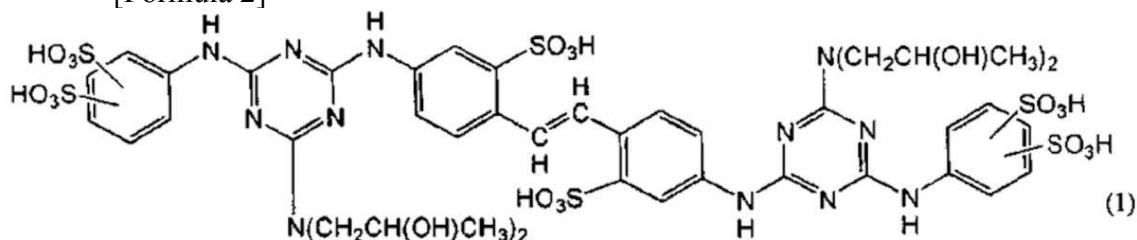


adding hydrochloric acid to the obtained reaction liquid to adjust the pH to 0.6, filtering the crystals that precipitated, and obtaining the compound expressed by formula (1); (H) adding 80 parts by mass of water to 50 parts by mass of these crystals that contain water, adjusting the pH to 8.5 by sodium hydroxide, and obtaining an aqueous solution containing 15 mass% of sodium salt of the compound expressed by formula (1);

[0025] (I) using 100 parts of ultrafine powder of silica (product of MIZUSAWA INDUSTRIAL CHEMICALS, LTD., trade name: MIZUKASIL P-78A), 30 parts of polyvinyl alcohol (product of Nippon Synthetic Chemical Industry Co., Ltd., trade name: GOHSENOL NM-11), 10 parts in terms of a solid content of polydiallyl dimethyl ammonium chloride that is a cationic dye fixing agent (quaternary ammonium salt of polymer) (product of SENKA corporation, trade name: HP-126A), and 0.9 parts in terms of the net content of the fluorescent brightener of an aqueous solution of a sodium salt of the compound expressed by formula (1); and adjusting the coating liquid concentration (solid content) so as to become 13%, by water.

Formula (1):

[Formula 2] "



#### B. Cited Document 1B invention

In paragraph [0014] of Cited Document 1, a size press coating liquid by an external addition method is disclosed, and the size press coating liquid contains "the fluorescent brightener expressed by the formula (1) or an aqueous solution thereof".

Then, in Cited Document 1, the following invention (hereinafter referred to as "Cited Document 1B invention") is described.

"[0014] A size press coating liquid contains a fluorescent brightener and an ink fixing agent;

[0007] the fluorescent brightener expressed by formula (1) is obtained by the steps of making a compound which is obtained by a step of making 2 moles of aniline disulfonic acid react with 2 moles of cyanuric chloride react with 1 mole of 4,4'-diaminostilbene-2,2'-disulfonic acid, and making the obtained compound react with 2 moles of di-2-propanolamine; and

[0021] the size press coating liquid produces ink jet archival paper that has high ink absorbability and high whiteness, by concomitantly using a cationic ink fixing agent in the size press by the external addition method.

[0014] The size press coating liquid.

Formula (1):

[Formula 2]



defects that the ink fixing agent and the fluorescent brightener are combined in the solution thereby to become insoluble in water, and the crystal precipitates, and that such a phenomenon occurs that required high whiteness fails to be obtained in dyeing.

[0006] As has been described above, there cannot be obtained any of stable thick aqueous solutions which contain a stilbene derivative that is used for brightening cellulose such as paper, pulp, or cotton with the fluorescent brightener, as the fluorescent brightener, unless the production process includes additional steps of desalting and the like.

[0007]

[Problem to be solved by the invention] In dyeing of cellulose such as paper, pulp, or cotton with the fluorescent brightener, it has been desired to develop a liquid composition which is excellent in storage stability at low temperature and high temperature and which is also excellent in a brightening effect, and to propose a method for fluorescent brightening the archival paper for an ink jet printer, which is excellent in fixability of the ink, water resistance, and whiteness degree."

D. "[0014]

[Embodiment of the invention] The water-borne liquid composition and the aqueous solution composition of the present invention will be described in detail below. The structures of the fluorescent brighteners of compound formula (1) and compound formula (2) have already been disclosed in Belgium Patent No. 719065.

[0015] The fluorescent brighteners expressed by formulae (1) and (2) have high stability in an aqueous solution, and can be prepared into a stable water-borne liquid composition and a stable aqueous solution composition, without removal of an inorganic salt and addition of alkanolamines, a quaternary ammonium salt, and the like, which become necessary when a water-borne liquid composition is prepared with the use of a fluorescent brightener having another, similar structure. In formulae (1) and (2), a cation corresponding to the sulfonic group represents any of cations of an alkali metal and an alkaline earth metal, or an ammonium ion, and a lithium ion or a sodium ion is preferable.

[0016] The concentration of the fluorescent brightener expressed by formula (1) in the water-borne liquid composition of the present invention is 10 to 40 mass%, and preferably is approximately 10 to 20 mass%."

E. "[0021] In addition, a content of the inorganic salt such as sodium chloride and sodium sulfate in the water-borne liquid composition and the aqueous solution composition of the present invention is 1.1 to 10 mass%. A rough indication of the content of the inorganic salt is indicated by the total content of the sodium chloride and sodium sulfate. A content of the sodium chloride is an amount obtained by measuring an amount of chlorine anions with titration by silver nitrate or with ion chromatography, and converting the value into a value of sodium chloride. A content of the sodium sulfate is an amount obtained by measuring an amount of sulphate anions with ion chromatography, and converting the value into a value of the sodium sulphate. When the content of the inorganic salt in the water-borne liquid composition and the aqueous solution composition exceeds 10 mass%, a crystal of the dye starts precipitation, and accordingly the storage stability becomes poor, which is not preferable, but as long as the content of the inorganic salt is in the above-described range, the water-borne liquid

composition and the aqueous solution composition of the present invention are highly stable and do not precipitate crystals even when the inorganic salt exists in the compositions. "

F. "[0022] The water-borne liquid composition and the aqueous solution composition of the present invention are suitable for dyeing of the cellulose-based material. The cellulose-based materials include paper, pulp, cotton, and the like; a coloring method for paper and pulp out of the above cellulose-based materials is roughly classified into an internal addition method of adding a fluorescent brightener to a pulp slurry in a step after the beating of the pulp and before the papermaking, and an external addition method of adding the fluorescent brightener to a size pressing liquid in a size pressing step after papermaking; but there are also methods of overcoating a coating liquid which has been prepared from the fluorescent brightener, an inorganic white pigment, a binder, and the like, onto the surface of the paper, and the like. The water-borne liquid composition and the aqueous solution composition of the present invention can be applied to any of the methods."

G. "[0024] In the size press by the external addition method, firstly, the pulp is beaten into a predetermined degree of beating by a pulper, a refiner, or the like, and is formed into a pulp slurry; usual filler, sizing agent, aluminum sulfate, fixing agent, and the like are further added appropriately; and then the mixture is subjected to papermaking according to an ordinary method. After that, a size press coating liquid is applied onto the paper, which contains the water-borne liquid composition and the aqueous solution composition of the present invention, with a size pressing machine that is arranged in a middle portion of a large number (usually, 20 to 60) of cylinder driers which are arranged in a process of drying paper with the cylinder drier; and then the coating liquid is dried. Thereby, the fluorescent-whitened paper is obtained. The above-described size press coating liquid is prepared by an appropriate mixture of the water-borne liquid composition and the aqueous solution composition of the present invention, starch, PVA, CMC, a surface sizing agent, water, and the like; and a content of the water-borne liquid composition and the aqueous solution composition in the size press coating liquid is usually 0.01 to 6.0% (net content); and an amount of the size press coating liquid to be coated is usually 0.5 to 3 g/m<sup>2</sup> (dry mass) of dried paper."

H. "[0026] The coating liquid for the overcoating by the external addition method on the archival paper having the receptive layer for ink jet can also be applied in a similar way. Firstly, the pulp is beaten into a predetermined degree of beating by a pulper, a refiner, or the like, and is formed into a pulp slurry; usual filler, sizing agent, aluminum sulfate, fixing agent, and the like are further added appropriately; and then the mixture is subjected to papermaking according to an ordinary method. A coat liquid (coating liquid) is prepared by the addition of water to a mixture which is formed of usually 5 to 30 parts by mass of an adhesive, 0.05 to 10 parts by mass of the fluorescent brightener, 0.1 to 0.5 parts by mass of a dispersing agent, and 0.50 to 30 parts by mass of a cationic ink fixing agent so that a solid matter in the mixture becomes usually 40 to 70 mass% with respect to 100 parts by mass of usually a white inorganic pigment; the coating liquid is applied onto the surface of the produced paper with a coater or a gate roll so that the coated amount becomes usually 5 to 40 g/m<sup>2</sup> (dry mass);

and the coating liquid is dried usually at 90 to 130°C, with a hot drier, for instance. Then, the fluorescent-whitened paper is obtained. This receptive layer can also have a layered structure formed of two or more layers. In addition, a paint for a gloss layer may also be applied onto the ink-receptive layer.

[0027] Usable cationic ink fixing agents include: higher fatty amine, a quaternary-ammonium-salt type compound, an ethylene oxide addition product of a secondary alkylamine, a cationic polymer compound, and inorganic particles whose surfaces are cationically charged."

I. "[0030] The water-borne liquid composition of the present invention is excellent in storage stability at low temperature and high temperature under the coexistence of an inorganic salt that has formed in synthesis and in a state of a synthesized sodium salt and the like, without being subjected to steps of removing the inorganic salt, converting the inorganic salt to a lower alkanolamine or a lower alkyl quaternary ammonium salt which enhance solution stability, and adding a solubilizing agent such as urea. In addition, the paper that has been fluorescent-whitened by the water-borne liquid composition of the present invention does not cause a quenching phenomenon; resists being affected by paper quality and a composition change of the size pressing liquid and the coat liquid; and exhibits excellent whiteness. Furthermore, excellent whiteness degree is obtained even when the water-borne liquid composition of the present invention is used together with a cationic ink fixing agent and a water resistant additive which are usually used in the archival paper for the ink jet printer."

J. "[0031]

[Examples] The present invention will be described further in detail below with reference to examples, but the present invention is not limited to these examples.

[0032] (Synthesis example and storage stability test of water-borne liquid composition)

Example 1

Twenty parts by mass of cyanuric chloride was dispersed with the use of 80 parts by mass of water, 40 parts by mass of ice and 0.1 parts by mass of a nonionic dispersing agent. To this dispersion liquid, 32.7 parts by mass of aniline-2,5-disulfonic acid was added. The dispersion liquid was cooled to a temperature of 5°C by an addition of ice, and the temperature was gradually raised from 5.0 to 25°C for 3 hours while the pH was kept at 4.0 to 4.5 by the addition of a sodium hydroxide solution. To this solution, an aqueous solution (130 parts by mass) was added dropwise for 3 hours, in which 19.5 parts by mass of 4,4'-diaminostilbene-2,2'-disulfonic acid was dissolved while the pH was adjusted so as to become 9.5 to 10.5 by sodium hydroxide. In the meantime, the sodium hydroxide solution was added to the solution so that the pH was kept at 4.0 to 4.5, and the temperature was gradually raised from 35 to 50°C. To this solution, 18 parts by mass of di-2-propanolamine were added, and the resultant solution was reacted at 80°C for 3 hours while the pH was kept at 8.5 to 9.0 by the addition of a sodium hydroxide solution. Hydrochloric acid was added to the obtained reaction liquid to adjust the pH to 0.6, the crystals that precipitated were filtered, and a free acid (M=H) of the compound expressed by formula (2) was obtained. To 50 parts by mass of these crystals containing water, water was added, the pH was adjusted to 8.5 by sodium hydroxide, and a water-borne liquid composition of the present invention was

obtained, which contained 15 mass% of a sodium salt of the compound expressed by formula (2). This composition contained 1.8 mass% of the inorganic salt, but the composition kept a stable dissolution state without precipitating a crystal of the fluorescent brightener, after the compositions had been stored at -5°C and 40°C for 2 months. ( $\lambda_{\text{max}}$  in water = 348 nm)

[0033] Example 2

The reaction liquid obtained in Example 1 was cooled to 50°C, crystals that were precipitated by the addition of 80 parts by mass of sodium chloride were filtered, and the sodium salt (M=Na) of the compound expressed by formula (2) was obtained. Water was added to 50 parts by mass of the crystals containing water, and a water-borne liquid composition of the present invention was obtained, which contained 20 mass% of the sodium salt of the compound expressed by formula (2). This composition contained 2.0 mass% of the inorganic salt, but the composition kept the stable dissolution state without precipitating the crystal of the fluorescent brightener, after the compositions were stored at -5°C and 40°C for 2 months.

[0034] Example 3

The reaction liquid obtained in Example 1 contains 15 mass% of the sodium salt (M=Na) of the compound expressed by formula (2). This reaction liquid contained 4.5 mass% of the inorganic salt, but the reaction liquid kept the stable dissolution state without precipitating the crystal of the fluorescent brightener, after the reaction liquids were stored at -5°C and 40°C for 2 months."

K. "[0040] (Example of dyeing: overcoating by external addition method)  
Example 4

There was produced a coat liquid which was adjusted so that the solid matter became 55%, by a step of adding water to a mixture formed of 20 parts of the water-borne liquid composition obtained in Example 1, 800 parts of clay, 200 parts of calcium bicarbonate, 3 parts of an acrylic dispersing agent (Kayacryl Resin C-220N, product of Nippon Kayaku Co., Ltd.), 50 parts of phosphorylated starch (MS-4600, product of NIHON SHOKUJIN KAKO CO., LTD.), 120 parts of latex (styrene butadiene copolymer L-1622 product of Asahi Chemical Industry Co., Ltd.), and 4 parts of a water resistant additive (Sumirez Resin 636, product of Sumitomo Chemical Company Limited); the coat liquid was applied onto high grade paper; the coat liquid was dried at 120°C; and the color was measured with the use of a spectrum whiteness colorimeter (SC-10W: product of Suga Test Instruments Co., Ltd.). The results are shown in Table-1. In addition, the whiteness degree was determined according to JIS P 8148."

L. "[0045] (Example of dyeing: size press by external addition method)  
Example 5

A liquid was prepared as a size coating liquid, in which 4 parts of an anionic surface sizing agent (Polymaron 382, product of ARAKAWA CHEMICAL INDUSTRIES, LTD.) was added to an aqueous solution formed of 20 parts of a water-borne liquid composition obtained in Example 1 and 976 parts of 3% oxidized starch (MS-3800, product of NIHON SHOKUJIN KAKO CO., LTD.). This liquid was sent to a size pressing machine, was applied onto weak sized paper having a Stockigt sizing degree of 7 seconds, and was dried at 65°C to 70°C. The color of

thereby fluorescent-whitened paper was measured with the use of a spectrum whiteness degree colorimeter (SC-10W: product of Suga Test Instruments Co., Ltd.). The results are shown in Table-2."

M. "[0050] (Overcoating by external addition method with the use of coating liquid for receptive layer for ink jet archival paper)

Example 6

A coating liquid for an receptive layer was prepared with the use of 100 parts of ultrafine powder silica (product of MIZUSAWA INDUSTRIAL CHEMICALS, LTD., trade name: MIZUKASIL P-78A), 30 parts of polyvinyl alcohol (product of Nippon Synthetic Chemical Industry Co., Ltd., trade name: Gosenol NM-11), 10 parts in terms of solid content of polydiallyl dimethyl ammonium chloride (quaternary ammonium salt of polymer) (product of SENKA corporation, trade name: HP-126A) which is a cationic dye fixing agent, and 6 parts of the water-borne liquid composition obtained in Example 1 so that a concentration (solid content) of the coating liquid became 13%. This coating liquid for the ink-receptive layer was applied onto a sheet of commercial neutral PPC paper with a bar coater so that the dry mass became 5 g/m<sup>2</sup>, and was dried at 120°C to produce the fluorescent-whitened paper provided with a receptive layer; and the color was measured. The results are shown in Table-3."

N. "[0056]

[Effect of Invention] The water-borne liquid composition of the fluorescent brightener expressed by formula (1) is excellent in storage stability at low temperature and high temperature, without being subjected to steps of removing the inorganic salt, converting the inorganic salt to a lower alkanolamine and a lower alkyl quaternary ammonium salt which enhance solution stability, and adding a solubilizing agent such as urea. Specifically, the water-borne liquid composition of the present invention is a stable water-borne liquid composition that contains a compound expressed by formula (1) with high concentration, even under the coexistence of an inorganic salt that has formed in synthesis, and is stable over a long time of period. The water-borne liquid composition of the present invention can be easily produced as compared to a conventional fluorescent brightener that contains a stilbene derivative with high concentration; and exhibits an excellent brightening effect in a method for fluorescent brightening cellulose such as paper, pulp, and cotton; is excellent in a fluorescent brightening force even when being used particularly together with a quaternary ammonium salt of a polymer and the like, which is an ink fixing agent that is used in ink-receptive paper; and is an extremely useful fluorescent brightener. Accordingly, the industrial value is extremely high."

(4) Invention described in Cited Document 2

A. Cited Document 2A invention

In paragraph [0032] of Cited Document 2, Example 1 is disclosed as an example of the water-borne liquid composition described in [Claim 2]. In addition, in paragraph [0050] of Cited Document 2, Example 6 is disclosed as an example of the overcoating by the external addition method on the ink jet archival paper, which uses the coating liquid for the receptive layer, which includes the water-borne liquid composition of Example 1.

Then, in Cited Document 2, the following invention (hereinafter referred to as "Cited Document 2A invention") is described.

"[0050] A coating liquid for a receptive layer of ink jet archival paper is a coating liquid for the receptive layer, which is used for overcoating by an external addition method on ink jet archival paper, and is prepared by the steps of:

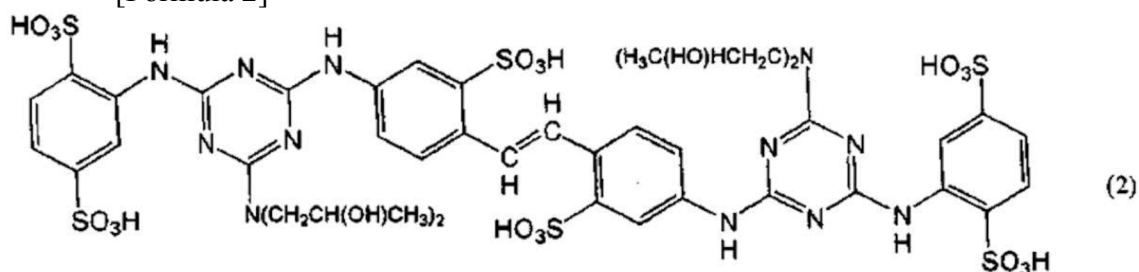
[0032] (A) dispersing 20 parts by mass of cyanuric chloride with the use of 80 parts by mass of water, 40 parts by mass of ice, and 0.1 parts by mass of a nonionic dispersing agent; (B) adding 32.7 parts by mass of aniline-2,5-disulfonic acid to this dispersion liquid; (C) cooling the dispersion liquid to a temperature of 5°C by adding ice, and raising the temperature of the dispersion liquid gradually from 5.0 to 25°C for 3 hours while keeping a pH at 4.0 to 4.5 by adding a sodium hydroxide solution; (D) adding the aqueous solution (130 parts by mass) to this solution dropwise for 3 hours, in which 19.5 parts by mass of 4,4'-diaminostilbene-2,2'-disulfonic acid is dissolved and the pH is adjusted so as to become 9.5 to 10.5 by sodium hydroxide; (E) in the meantime, adding a sodium hydroxide solution to the solution to keep the pH at 4.0 to 4.5, and gradually raising the temperature from 35 to 50°C; (F) adding 18 parts by mass of di-2-propanolamine to this solution, and making the compounds react at 80°C for 3 hours while keeping the pH at 8.5 to 9.0 by adding a sodium hydroxide solution; (G) adding hydrochloric acid to the obtained reaction liquid to adjust the pH to 0.6, filtering the crystals that precipitate, and obtaining a free acid (M=H) of the compound expressed by formula (2); (H) adding water to 50 parts by mass of these crystals containing water, and adjusting the pH to 8.5 by sodium hydroxide to obtain a water-borne liquid composition; and

[0050] (I) adjusting a concentration (solid content) of the coating liquid so as to become 13%, by using 100 parts ultrafine powder silica (product of MIZUSAWA INDUSTRIAL CHEMICALS, LTD., trade name: MIZUKASIL P-78A), 30 parts of polyvinyl alcohol (product of Nippon Synthetic Chemical Industry Co., Ltd., trade name: GOHSENOL NM-11), 10 parts in terms of solid content of polydiallyl dimethyl ammonium chloride (quaternary ammonium salt of polymer) (product of SENKA corporation, trade name: HP-126A) which is a cationic dye fixing agent, and 6 parts of the water-borne liquid composition.

Here, the water-borne liquid composition contains

[Claim 2] 15 mass% of a fluorescent brightener represented by the following formula (2) in a form of a free acid,

[Formula 2]



(In the formula, cation corresponding to sulfonic group is cation of sodium), wherein the content of the inorganic salt is 1.8 mass%."

B. Cited Document 2B invention



In paragraph [0024] of Cited Document 2, a size press coating liquid produced by an external addition method is disclosed, and the size press coating liquid contains "a water-borne liquid composition of the present invention".

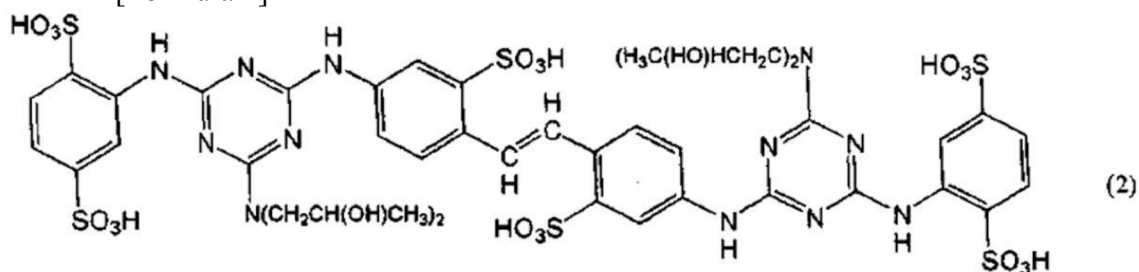
Then, in Cited Document 2, also the following invention (hereinafter referred to as "Cited Document 2B invention") is described.

"[0024] A size press coating liquid is a size press coating liquid that provides fluorescent-whitened paper by being applied onto a sheet of paper with a size pressing machine, and being dried, where the size press coating liquid is prepared from an appropriate mixture of the water-borne liquid composition, starch, PVA, CMC, a surface sizing agent, water, and the like, where

the water-borne liquid composition contains

[Claim 2] 10 to 40 mass% of a fluorescent brightener represented by the following formula (2) in a form of a free acid,

[Formula 2]



(In the formula, a cation corresponding to the sulfonic group is any of cations of an alkali metal and an alkaline earth metal, or an ammonium ion), wherein a content of an inorganic salt is 1.1 to 10 mass%."

## 2 Comparison to Cited Document 1A Invention and Judgment

### (1) Comparison

#### A. Sizing composition

"A coating liquid for a receptive layer" of Cited Document 1A invention is "a coating liquid for a receptive layer for ink jet archival paper". Here, "the ink jet archival paper" of Cited Document 1A invention corresponds to "a substrate for ink jet printing" of the Invention. In addition, "a sizing composition" of the Invention is "applied" onto "a substrate for ink jet printing" (paragraph [0029] of detailed description of the Invention of the present application), and is "a liquid" in view of the composition. Accordingly, "a coating liquid for a receptive layer" of Cited Document 1A invention and "a sizing composition" of the Invention are common in the point of "the coating liquid". In addition, "the coating liquid for receptive layer" of Cited Document 1A invention contains "a fluorescent brightener", and accordingly is a substance that optically brightens "ink jet archival paper", from the technical viewpoint.

Accordingly, "the coating liquid for the receptive layer" of Cited Document 1A invention and "the sizing composition" of the Invention are common in the point of "the coating liquid that optically brightens the substrate for ink jet printing".

#### B. Binder

"A coating liquid for a receptive layer" of Cited Document 1A invention contains "polyvinyl alcohol", and the "polyvinyl alcohol" corresponds to "(a) at least one binder" of the Invention in consideration of technical knowledge.

### C. Water

"The coating liquid for the receptive layer" of Cited Document 1A invention is a coating liquid "in which 0.9 parts in terms of the net content of the fluorescent brightener of an aqueous solution of a sodium salt of the compound expressed by formula (1) has been adjusted by water so that a concentration (solid content) of the coating liquid becomes 13%", and accordingly contains "(c) water", similarly to the Invention.

### D. Optical brightener of Formula (1)

"The coating liquid for the receptive layer" of Cited Document 1A invention is a liquid "which uses 0.9 parts in terms of the net content of the fluorescent brightener of the aqueous solution of the sodium salt of the compound of formula (1)". In addition, "the compound expressed by formula (1)" of Cited Document 1A invention uses "aniline-2,5-disulfonic acid" as a starting substance.

Accordingly, "the sodium salt of the compound of formula (1)" of Cited Document 1A invention satisfies a requirement of "at least one optical brightener of formula (1)" of the Invention.

### (2) Corresponding features and the different features

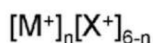
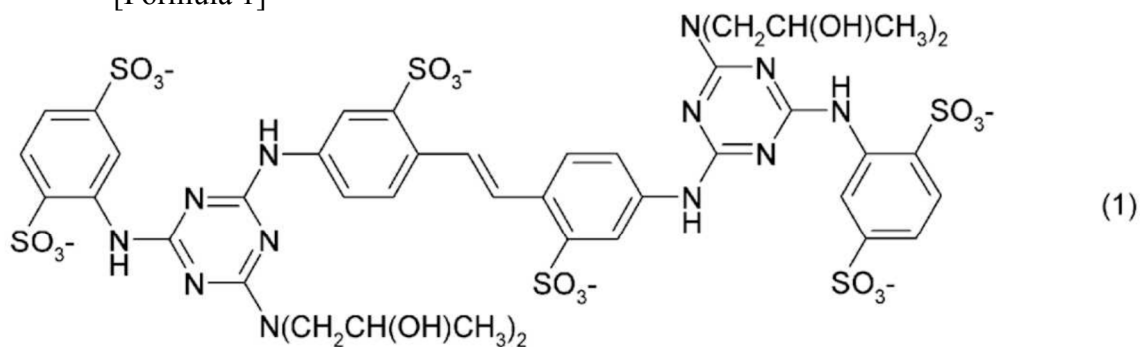
#### A. Corresponding features

The Invention and Cited Document 1A invention correspond with each other in the following constitution.

"A coating liquid for optically brightening a substrate for ink jet printing, including:

- (a) at least one binder;
- (c) water; and
- (d) at least one optical brightener of formula (1).

[Formula 1]



(In the formula,

M and X are identical or different and independently from each other selected from the group consisting of hydrogen, an alkali metal cation, ammonium, ammonium which is mono-, di- or trisubstituted by a C<sub>1</sub>-C<sub>4</sub> linear or branched alkyl radical, ammonium which is mono-, di- or trisubstituted by a C<sub>1</sub>-C<sub>4</sub> linear or branched hydroxyalkyl radical, or a mixture of said compounds, and n is in the range from 0 to

6.)"

B. The different features

The Invention and Cited Document 1A invention are different from each other, or are prima facie different from each other, in the following points.

(The different feature 1)

The point in which "a coating liquid" of the Invention includes "(b) at least one divalent metal salt being selected from the group consisting of calcium chloride, magnesium chloride, calcium bromide, magnesium bromide, calcium iodide, magnesium iodide, calcium nitrate, magnesium nitrate, calcium formate, magnesium formate, calcium acetate, magnesium acetate, calcium sulphate, magnesium sulphate, calcium thiosulphate or magnesium thiosulphate, or mixtures of said compounds", but on the other hand, Cited Document 1A invention does not include, in view of a production process, "(b) at least one divalent metal salt being selected from the group consisting of calcium chloride, magnesium chloride, calcium bromide, magnesium bromide, calcium iodide, magnesium iodide, calcium nitrate, magnesium nitrate, calcium formate, magnesium formate, calcium acetate, magnesium acetate, calcium sulphate, magnesium sulphate, calcium thiosulphate or magnesium thiosulphate, or mixtures of said compounds".

(The different feature 2)

The point in which the "coating liquid" of the Invention is "a sizing composition", but on the other hand, "the coating liquid" of Cited Document 1A invention is "a coating liquid for a receptive layer".

(3) Judgment

The judgment regarding the different feature 1 and the different feature 2 is described as follows.

A. Regarding the different feature 1

"A coating liquid for a receptive layer" of Cited Document 1A invention includes "polydiallyl dimethyl ammonium chloride (quaternary ammonium salt of polymer)" serving as "a cationic dye fixing agent", and "a divalent metal salt" of "calcium chloride" and the like is well known as a compound that functions as "the cationic dye fixing agent", as is described below.

(A) Japanese Unexamined Patent Application Publication No. 2004-50532 (well-known literature cited in notice of reasons for refusal)

"[0029]

In the present invention, in order to enhance the fixability of printed ink and improve water resistance, it is preferable that the ink-receptive layer contains a cationic polymer or a polyvalent metal. Particularly when a fine particle of silica or further silica by a vapor-phase method is used as an inorganic fine particle, it is preferable that the ink-receptive layer contains a cationic polymer or a polyvalent metal. The cationic polymer or the polyvalent metal can be added to the coating liquid for the ink-receptive layer in arbitrary timing, but is preferably mixed with a dispersion liquid of the inorganic fine particle before a mixture solution of a PVA-fluorescent brightener is mixed with the dispersion liquid of the inorganic fine particle. It is particularly preferable to disperse the inorganic fine particles in the presence of the cationic polymer

or the polyvalent metal, from the viewpoint of stabilizing the dispersion of the inorganic fine particles and fixing an ink dye."

"[0031]

Polyvalent metals which are preferably used in the present invention include calcium, barium, manganese, copper, cobalt, nickel, aluminum, iron, zinc, zirconium, titanium, chromium, magnesium, tungsten and molybdenum; and these metals can be used in the form of water-soluble salts. Specifically, the water-soluble salts include, for instance, calcium acetate, calcium chloride, calcium formate, and calcium sulfate ... (omitted)."

(B) Japanese Unexamined Patent Application Publication No. 2004-50501 (well-known literature cited in notice of reasons for refusal)

"[0079]

<Mordant>

In the present invention, in order to enhance water resistance of a formed image and temporal bleeding resistance, a mordant may be contained in a coloring material receptive layer. Incidentally, a cation polymer of general formula (1) also has a function of the mordant.

The above-described mordant is preferably a cationic polymer (cationic mordant) that is an organic mordant, or an inorganic mordant; and by existing in the coloring material receptive layer, the mordant interacts with a liquid ink having an anionic dye as a coloring material, stabilizes the coloring material, and can enhance water resistance and temporal bleeding resistance. The organic mordant and the inorganic mordant may be used independently, and the organic mordant and the inorganic mordant may be used in combination."

"[0091]

Examples of the above-described inorganic mordant include a polyvalent water-soluble metal salt and a hydrophobic metal salt compound.

Specific examples of the inorganic mordant include, for instance, a salt or a complex of a metal selected from the group consisting of magnesium, aluminum, calcium, scandium, titanium, vanadium, manganese, iron, nickel, copper, zinc, gallium, germanium, strontium, yttrium, zirconium, molybdenum, indium, barium, lanthanum, cerium, praseodymium, neodymium, samarium, europium, gadolinium, dysprosium, erbium, ytterbium, hafnium, tungsten, and bismuth.

[0092]

Specifically, the inorganic mordants include, for instance, calcium acetate, calcium chloride, calcium formate, and calcium sulfate ... (omitted)."

(C) Japanese Unexamined Patent Application Publication No. H9-230534 (well-known literature cited in decision of refusal)

"[0064] Example 1

(Base paper A) ... (omitted) ... a stiff slurry was put on a long net paper machine that runs at 200 m/minute; a web was formed while an appropriate turbulence was imparted; the formed web was subjected to three-stage wet press whose line pressure was adjusted in a range of 15 kg/cm to 100 kg/cm at a wet part, then was

processed by a smoothing roll, and was subsequently subjected to a two-stage bulk density press whose line pressure was adjusted in a range of 30 kg/cm to 70 kg/cm at a dried part; and the web was dried. After that, a size pressing liquid was applied onto the web in an amount of 25 g/m<sup>2</sup> on the way of drying, which was formed of 4 parts by weight of a carboxy modified polyvinyl alcohol, 0.05 parts by weight of a fluorescent brightener, 0.002 parts by weight of a blue dye, 4 parts by weight of sodium chloride, and 92 parts by weight of water; the web was size pressed; and the web was dried so that the water content of the base paper to be finally obtained became 8 wt.% in terms of absolute dry water content, and was subjected to a machine calendar process of which the line pressure was 70 kg/cm. Thus, there was produced base paper A whose basis weight was 170 g/m<sup>2</sup> and density was 1.04 g/cm<sup>3</sup>."

"[0069] (Base paper F) There was produced base paper F whose basis weight was 170 g/m<sup>2</sup> and density was 1.06 g/cm<sup>3</sup>, in a similar way to the base paper A, except that in place of a stiff slurry used in the base paper A, there was used a stiff slurry which was prepared by beating a bleached kraft pulp of a broad-leaved tree so that a fiber length of the pulp after having beaten became 0.51 mm (when indicated by weighted mean fiber length of lengths that were measured according to JAPAN TAPPI paper pulp testing method No. 52-89 "testing method of paper and pulp fiber length"), and so that Canadian standard freeness became 270 ml, and by adding 0.6 parts by weight (as solid content) of polyaluminum hydroxide, 1.5 parts by weight of cationized starch, 0.2 parts by weight of anionized polyacrylamide, 1.5 parts by weight of sodium stearate, 0.1 parts by weight of epoxidized higher fatty acid amide, 0.4 parts by weight of a polyamide epichlorohydrin resin and an appropriate amount of a fluorescent brightener, a blue dye and a red dye, with respect to 100 parts by weight of the pulp, and also the same except that 4 parts by weight of calcium chloride was used in place of 4 parts by weight of sodium chloride in the size pressing liquid."

(D) Japanese Unexamined Patent Application Publication No. 2003-326841

"[0024] In the present invention, the kraft pulp formed from acacia is beaten by a disk refiner or the like so that a degree of water retention obtained from the above-described formula becomes 150 to 180%. On the other hand, a pulp other than the kraft pulp formed from acacia is separately prepared, and the prepared pulp is mixed with the above-described kraft pulp formed from acacia. In a range in which the degree of the water retention of the kraft pulp formed from acacia is 150 to 180%, a pulp fiber is sufficiently flexible, an effect of improving smoothness is achieved, poor dehydration does not occur on the paper machine wire, and the amount of dry steam to be used does not increase.

[0025] To the above-described mixed pulp, various compounds are added, as needed, which include: a filler such as clay, talc, calcium carbonate, or fine particles of a urea resin; a sizing agent such as rosin, an alkyl ketene dimer, a higher fatty acid, an epoxidized fatty acid amide, paraffin wax, or alkenyl succinic acid; a paper strengthening agent such as starch, polyamide polyamine epichlorohydrin, or polyacrylamide; and a fixing agent such as aluminum sulfate or a cationic polymer. Then, the resultant mixture is used.

[0026] The slurry of the pulp, which has been prepared in the above-described way, is subjected to paper making. This paper making step includes a step of drying a

web face side that corresponds to a surface of body paper to be coated with a photographic emulsion, while pressing a drier canvas onto a drum drier cylinder; and in this step, a tension force of the drier canvas is adjusted in a range of 1.5 to 3 kg/cm, and the web face side is dried.

[0027] The surface of the body paper (one side or both sides thereof) which has been dried in the above way can be coated with: poly vinyl alcohol or its modified product, and/or starch; a fluorescent brightener such as diaminostilbene disulfonic acid; and a chloride of a polyvalent metal such as calcium chloride, magnesium chloride, and aluminum chloride."

(E) Japanese Unexamined Patent Application Publication No. H2-118200

"(Field of industrial application)

The present invention relates to a paper making method having a surface sizing step." (lines 10 to 12 in left column on page 1)

"(Example)

The paper making method of the present invention was carried out on the following common conditions.

A long strip of paper was subjected to surface sizing by a pass line in the vicinity of a size press illustrated in Figure 1, and then to paper making. As for a driving roll G whose surface temperature is variable, there was used the roll illustrated in Figures 2 to 3. The surface size liquid that was used is formed of polyvinyl alcohol, calcium chloride, a fluorescent brightener, a defoaming agent, gelatin, and an antiseptic agent." (lines 12 to 20 in left column on page 3)

(F) Japanese Unexamined Patent Application Publication No. 2005-313454

"[0050]

In addition, a metal salt can be used as the cationic substance, in addition to the above-described organic polymer. For instance, there is known a technology that treats the surface of the paper with a salt of a metal which includes magnesium, calcium, and aluminum, and enhances an image quality (for instance, see Patent document 8 and the like).

Also in the present invention, fundamentally, one or more known metal salts can be used, and a metal salt containing a divalent or polyvalent metal ion is preferable. It is more preferable to use one or more of calcium, magnesium, strontium, barium, and radium as the divalent or polyvalent metal ion. It is further preferable to use one or more of calcium and magnesium."

"[0145]

<Example 2>

A kraft pulp of a broad-leaved tree was subjected to bleaching treatment by a TCF multistage-bleaching method that includes a xylanase treatment step, an alkaline extraction step, a hydrogen peroxide treatment step, and an ozonation step. The obtained pulp was beaten and adjusted so that a source water degree became 450 ml; the beaten pulp was blended with 3 parts by mass of a kaolin filler, 6 parts by mass of a filler of precipitated calcium carbonate light, and 0.2 parts by mass of an internal sizing agent of an alkenyl succinic anhydride (ASA), with respect to 100 parts by mass of the

above described pulp; and the mixture was subjected to paper making. Furthermore, a coating liquid (viscosity of coating liquid; 25 mPa-s (at liquid temperature of 60°C)) was prepared that was formed of 98 parts by mass of water, 1 part by mass of polyacrylic acid, and 1 part by mass of a quaternary ammonium salt (trade name: HP200A (product of SENKA corporation), cation equivalent; 4.3 meq/g) as a surface sizing agent; and the paper was subjected to size pressing. Then, archival paper was obtained whose surface was coated with 0.5 g/m<sup>2</sup> of polyacrylic acid and 0.5 g/m<sup>2</sup> of the quaternary ammonium salt. The electro-conductivity was 0.0025 S/m.

[0146]

<Example 3>

A mechanical pulp of a needle-leaved tree was subjected to bleaching treatment with hydrosulfite, and was beaten and adjusted so that the freeness became 450 ml; and the beaten pulp was blended with 8 parts by mass of a light calcium carbonate filler, and 0.02 parts by mass of an internal sizing agent of alkenyl succinic anhydride (ASA), with respect to 100 parts by mass of the previously described pulp. Furthermore, there was prepared a coating liquid (viscosity of coating liquid; 20 mPa-s (at liquid temperature of 60°C)) that was formed of 94 parts by mass of water, 1 part by mass of a cation modified polyvinyl alcohol (product of Nippon Synthetic Chemical Industry Co., Ltd., GOHSEFIMER K210), and 5 parts by mass of calcium bromide as a surface sizing agent; and the paper was subjected to size pressing. The surface of the obtained archival paper was coated with 1.0 g/m<sup>2</sup> of calcium bromide and 0.2 g/m<sup>2</sup> of cation modified polyvinyl alcohol. The electro-conductivity was 0.0145 S/m."

In addition to the above-described well-known art, in paragraphs [0002] and [0003] of Cited Document 1, the following are respectively described: "[Prior art] ...(omitted)... anionic dye is used in the ink of an ink jet printer using a water-soluble dye. Because of this, in archival paper for an ink jet printer of high quality, a cationic ink fixing agent, a water resistant additive, and the like are used in many cases when the ink jet archival paper is produced, in order to suppress the bleeding of ink on the paper. In addition, the archival paper is required to have clear color developability for an image, and accordingly is also required to have whiteness of high quality. Because of this, there are produced sheets of ink jet archival paper which are dyed into high whiteness with the use of various fluorescent brighteners. As for the fluorescent brightener that is used at this time, in many cases the anionic water-soluble fluorescent brightener is used, from the viewpoint of characteristics in the production process. However, when the above-described cationic fixing agent and the anionic fluorescent brightener are used at the same time, there have been significant problems that the ink fixing agent and the fluorescent brightener are combined in the solution to thereby become insoluble in water, and the crystal precipitates, and that there occurs such a phenomenon that high whiteness required in dyeing is not obtained.", and "[Problem to be solved by the invention] An object of the present invention is to solve the above described problems, and to provide a method for producing receptive paper for the ink jet, which has whiteness of high quality, while simultaneously using a cationic ink fixing agent and an anionic water-soluble fluorescent brightener." In addition, such knowledge is disclosed that a fluorescent brightener represented by formula (1) of Cited Document 1A invention is excellent as means for solving the problem.

In consideration of the above description, it is a matter in a range of usual originality and creativity for a person skilled in the art to independently or concomitantly use the divalent metal salt concerning the different feature 1, as the cationic ink fixing agent of Cited Document 1A invention.

#### B. Regarding the different feature 2

Regarding the term of "a sizing composition", the definition is not described in the detailed description of the Invention of the present application, but "size" usually means "a chemical which is added to paper stuff or is applied onto a paper surface when the paper is produced, in order to prevent bleeding of ink on the paper and fuzzing of the paper: rosin, casein, gelatin, starch, a synthetic resin, and the like." (the 6th edition of Kojien). Accordingly, a film is formed on the paper surface depending on the type, the viscosity, and the like of the chemical (sizing composition) to be applied, and the film is not substantially distinguished from the coating. Then, the Invention includes "(a) at least one binder", but in the detailed description of the Invention of the present application, it is described that "one or more secondary binders such as polyvinyl alcohol may be used" (paragraph [0018]), and a material is also disclosed which has strong film formability and is often used in a coating agent for paper.

In addition, in the detailed description of the Invention of the present application, it is described that "the sizing composition may be applied onto the surface of the paper substrate with an arbitrary surface treatment method which is well known in the technical field; and examples of the application method include size press application, calendar size application, tab sizing, coating application, and spray application" (paragraph [0029]), and it is disclosed that "application" is not limited to "sizing", and may also be "coating".

Furthermore, in the detailed description of the Invention of the present application, it is described that "the sizing composition may contain a by-product that is formed in preparation of an optical brightener, and other conventional paper additives; and examples of such an additive are a carrier, a defoaming agent, a wax emulsion, a dye, an inorganic salt, a solubilizing auxiliary agent, an antiseptic agent, a complexing agent, a surface sizing agent, a cross linking agent, a pigment, a special resin, and the like" (paragraph [0025]). Here, regarding the term of "the pigment", the definition is not described in the detailed description of the Invention of the present application, but in paragraph [0006], "pigment (preferably, any one of precipitated calcium carbonate and calcium carbonate heavy)" is described, and exemplification of the specific substances other than the above description does not exist. The precipitated calcium carbonate and the calcium carbonate heavy are typical materials serving as a receptive layer material in overcoating of the ink jet printing paper. For confirmation, reference is made to Patent Application Laid-Open No. 2009-513843, which is International Publication No. WO 2007-053681 (applicant: International Paper company), and there, the following are described in paragraph [0006].

(A) "[0002]

#### Field of invention

The present invention relates to a sizing composition that, when applied to a paper substrate, creates a substrate, preferably suitable for inkjet printing, having increased print density, print sharpness, low HST, and/or image dry time, the substrate preferably having high whiteness and reduced color-to-color bleed as well. In addition,



the present invention relates to a method of reducing the HST of a paper substrate by applying the sizing composition to at least one surface thereof. Further, the application relates to methods of making and using the sizing composition, as well as methods of making and using the paper containing the sizing composition."

(B) "[0006]

The sizing composition may contain a pigment. Examples of pigments are clay, calcium carbonate, calcium sulfate hemihydrate, and calcium sulfate dehydrate, calcium carbonate, preferably precipitated calcium carbonate, in any form including ground calcium carbonate and silica-treated calcium carbonate. When the pigment is a calcium carbonate, it may be in any form. Examples include ground calcium carbonate and/or precipitated calcium carbonate. Commercially available products that are preferred are those offered as Jetcoat 30 from Specialty Minerals Inc., Jetcoat MD 1093 from Specialty Minerals Inc., XC3310-1 from Omya Inc, and OmyaJet B5260, C4440 and 6606 from Omya Inc.

[0007]

The pigment may have any surface area. Those pigments having a high surface area are included, including those having a surface area of greater than 20 square meters/gram, preferably greater than 30 square meters/gram, more preferably greater than 50 square meters/gram, most preferably greater than 100 square meters/gram. This range includes greater than or equal to 1, 5, 10, 15, 20, 25, 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, 80, 85, 90, 100 square meters/gram, including any and all ranges and subranges contained therein.

[0008]

The sizing composition may contain a pigment in any amount. The composition may include pigment in an amount from 0 to 99 wt% based upon the total weight of the solids in the composition, preferably at least 15 wt%, more preferably at least 30 wt%, most preferably at least 45 wt%, based upon the total weight of the solids in the composition. This range may include 0, 1, 5, 10, 15, 20, 25, 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, 80, 85, 90, 100 wt% of pigment based upon the total weight of the solids in the composition, including any and all ranges and subranges contained therein. The most preferred amount is about 52 wt% pigment based upon the total weight of the solids in the composition."

(C) "[0016]

The sizing composition may contain at least one inorganic salt. Suitable inorganic salts may be monovalent and/or divalent and/or trivalent and may contain any level of hydration complexes thereof. Exemplified inorganic salts are those from Groups 1, 2 and 13 from the Periodic Table of Elements and hydrated complexes thereof, including monohydrates, dihydrates, trihydrates, tetrahydrates, etc. The cationic metal may preferably be sodium, calcium, magnesium, and aluminum. The anionic counterion to the cationic metal of the inorganic salt may be any halogen, such as chloride, boride, fluoride, etc., and/or hydroxyl group(s). The most preferred inorganic salt is sodium chloride."

(D) "[0033]

Figures 1-3 demonstrate different embodiments of the paper substrate 1 in the

paper substrate of the present invention. Figure 1 demonstrates a paper substrate 1 that has a web of cellulose fibers 3 and a sizing composition 2, where the sizing composition 2 has minimal interpenetration of the web of cellulose fibers 3. Such an embodiment may be made, for example, when a sizing composition is coated onto a web of cellulose fibers.

[0034]

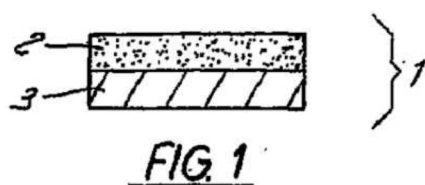
Figure 2 demonstrates a paper substrate 1 that has a web of cellulose fibers 3 and a sizing composition 2 where the sizing composition 2 interpenetrates the web of cellulose fibers 3. The interpenetration layer 4 of the paper substrate 1 defines a region in which at least the sizing solution penetrates into and is among the cellulose fibers. The interpenetration layer may be from 1 to 99% of the entire cross section of at least a portion of the paper substrate, including 1, 2, 5, 10, 15, 20, 25, 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, 80, 85, 90, 95, and 99% of the paper substrate, including any and all ranges and subranges therein. Such an embodiment may be made, for example, when a sizing composition is added to the cellulose fibers prior to a coating method and may be combined with a subsequent coating method if required. Addition points may be at the size press, for example.

[0035]

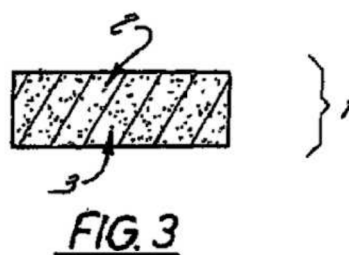
Figure 3 demonstrates a paper substrate 1 that has a web of cellulose fibers 3 and a sizing solution 2 where the sizing composition 2 is approximately evenly distributed throughout the web of cellulose fibers 3. Such an embodiment may be made, for example, when a sizing composition is added to the cellulose fibers prior to a coating method and may be combined with a subsequent coating method if required. Exemplified addition points may be at the wet end of the paper making process, the thin stock, and the thick stock."

[Figure 1] to [Figure 3]

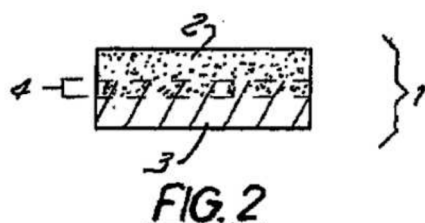
【図 1】



【図 3】



【図 2】



When the above-described circumstance is taken into consideration, it should be understood that "the sizing composition" in the Invention is "the sizing composition" which includes the application to be used as "the coating liquid for the receptive layer for the overcoating by the external addition method on the ink jet archival paper".

Accordingly, the different feature 2 is not a substantial different feature.

Or, the size press coating liquid is well known without pausing to exemplify, which contains the optical brightener and the cationic ink fixing agent. Incidentally, in paragraph [0021] of Cited Document 1, it is described that "The compound expressed by formula (1) in the present invention can produce ink jet archival paper of high whiteness by being used in a size press by the internal addition method and the external addition method, in overcoating by the external addition method, and in the ink-receptive layer. Among these uses, in the size press by the external addition method, the overcoating by the external addition method, and the ink-receptive layer, there can also be produced ink jet archival paper that has high ink absorbability and high whiteness, by concomitantly using a cationic ink fixing agent." In addition, it is self-evident that the size press coating liquid for the external addition method contains "(a) at least one binder" and "(c) water". Incidentally, also in paragraph [0014] of Cited Document 1, it is described that "the size press coating liquid is prepared from an appropriate mixture of starch, PVA, CMC, a surface sizing agent, water, and the like" (Also in paragraph [0018] of the present application, starch (starch) and polyvinyl alcohol (PVA) are exemplified as a binder.). Furthermore, such a point is, as has been determined in the different feature 1, that the cationic ink fixing agent is "(b) at least one divalent metal salt being selected from the group consisting of calcium chloride, magnesium chloride, calcium bromide, magnesium bromide, calcium iodide, magnesium iodide, calcium nitrate, magnesium nitrate, calcium formate, magnesium formate, calcium acetate, magnesium acetate, calcium sulphate, magnesium sulphate, calcium thiosulphate or magnesium thiosulphate, or mixtures of said compounds".

Accordingly, in Cited Document 1A invention, it is a matter in a range of usual originality and creativity for a person skilled in the art to prevail the different feature 2.

#### (4) Effect of the Invention

In the detailed description of the Invention of the present application, there is no explicit description which describes the effect of the Invention, but in the detailed description of the Invention of the present application, the following are described.

A. "[Problem to be solved by the invention]

[0010]

Accordingly, there is a need for a water-soluble optical brightener which has good compatibility with sizing compositions containing a divalent metal salt.

Description of the invention

[0011]

It has now been found that optical brighteners of formula (1) have surprisingly good compatibility with sizing compositions containing a divalent metal salt."

B. "[0064]

The results in Table 1 clearly demonstrate the excellent whitening effect afforded by the compositions of the invention."

C. "[0068]

The results in Table 2 show that the composition of the invention has no adverse effect on ink print density."

In view of the descriptions of [Prior Art] (paragraph [0002]), [Problem to be solved by the invention] (paragraph [0003]), and [Means for solving problem] (paragraphs [0004] to [0006]) of Cited Document 1, the problem and the solving means of the above described A. are only matters that are also disclosed in Cited Document 1.

In addition, in paragraph [0022] of Cited Document 1, it is described that "ink jet archival paper using a compound of formula (1) has whiteness of high quality, and also is excellent in adequate ink absorbency and printing characteristics that a clear image quality free from no bleeding of ink is obtained", and accordingly the above described effects of B. and C. are merely effects which Cited Document 1A invention shows or are merely, at least, an effect which a person skilled in the art having contacted Cited Document 1 expects.

#### (5) Summary

The Invention is an invention which a person skilled in the art would have easily made based on Cited Document 1A invention and the well-known art.

### 3 Comparison to Cited Document 1B Invention and Judgment

#### (1) Comparison

When the Invention and Cited Document 1B invention are compared to each other, the result is as follows.

##### A. Sizing composition

The "size press coating liquid" of Cited Document 1B invention is a "size press coating liquid that contains a fluorescent brightener and an ink fixing agent", and is a liquid that "produces ink jet archival paper that has high ink absorbability and high whiteness by being used together with a cationic ink fixing agent in the size press by the external addition method."

Here, the "ink jet archival paper" and the "size press coating liquid" of Cited Document 1B invention correspond to the "substrate for ink jet printing" and the "sizing composition" of the Invention, respectively. In addition, the "size press coating liquid" of Cited Document 1B invention includes the "fluorescent brightener", and accordingly is a substance that optically brightens "ink jet archival paper", from the technical viewpoint

Accordingly, the "size press coating liquid" of Cited Document 1B invention satisfies a requirement of the "sizing composition that optically brightens the substrate for ink jet printing" of the Invention.

##### B. Optical brightener of formula (1)

The "fluorescent brightener of formula (1)" of Cited Document 1B invention includes the invention in which the "cation corresponding to sulfonic group" is the "cation of an alkali metal".

Then, the "fluorescent brightener of formula (1)" of Cited Document 1B invention satisfies a requirement of the "optical brightener of formula (1)" of the Invention, except that a position at which the sulfonic group is bonded is not specified.

#### (2) Corresponding features and the different features

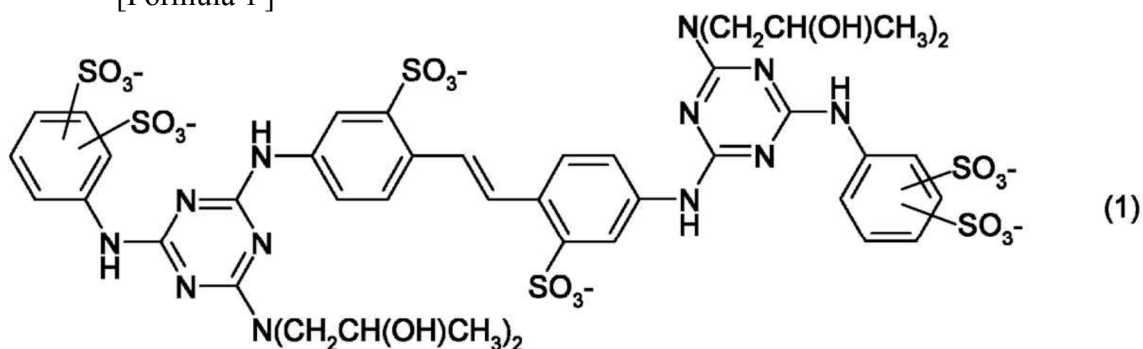
##### A. Corresponding features

The Invention and Cited Document 1B invention correspond with each other in the following constitution.

"A sizing composition that optically brightens a substrate for ink jet printing, comprising:

(d) at least one optical brightener of formula (1).

[Formula 1']



(In the formula,

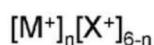
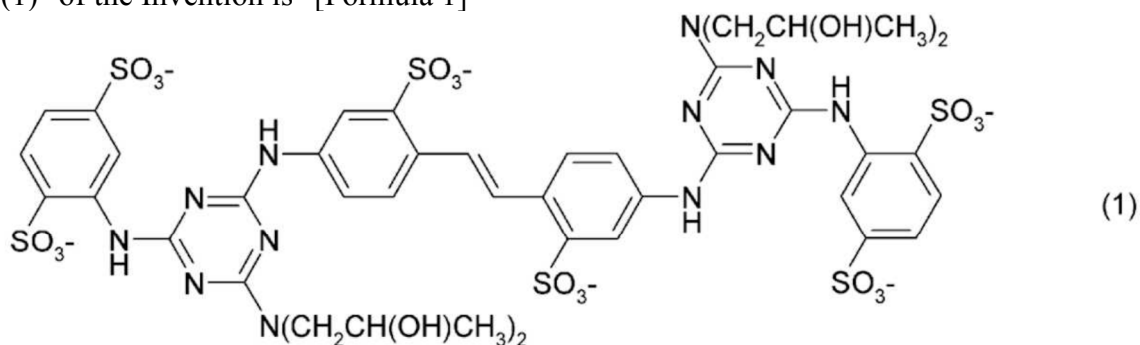
M and X are identical or different, and independently from each other selected from the group consisting of hydrogen, an alkali metal cation, ammonium, ammonium which is mono-, di- or trisubstituted by a C<sub>1</sub>-C<sub>4</sub> linear or branched alkyl radical, ammonium which is mono-, di- or trisubstituted by a C<sub>1</sub>-C<sub>4</sub> linear or branched hydroxyalkyl radical, or mixtures of said compounds, and n is in the range from 0 to 6.)"

B. The different features

The Invention and Cited Document 1B invention are different from each other, or are prima facie different from each other, in the following points.

(The different feature 3)

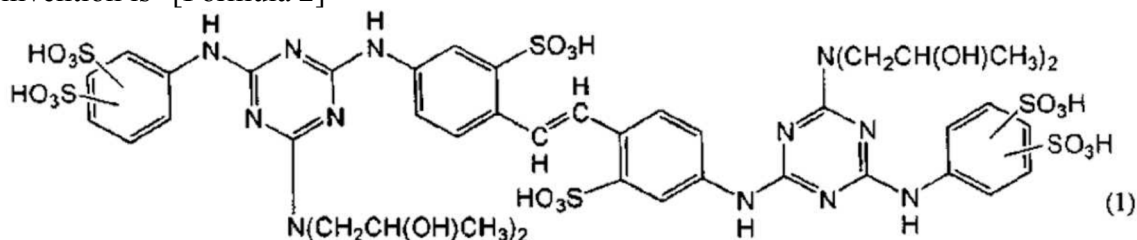
The point in which a structural formula of the "optical brightener of formula (1)" of the Invention is "[Formula 1]



(In the formula,

M and X are identical or different, and independently from each other selected from the group consisting of hydrogen, an alkali metal cation, ammonium, ammonium

which is mono-, di- or trisubstituted by a C<sub>1</sub>-C<sub>4</sub> linear or branched alkyl radical, ammonium which is mono-, di- or trisubstituted by a C<sub>1</sub>-C<sub>4</sub> linear or branched hydroxyalkyl radical, or mixtures of said compounds, and n is in the range from 0 to 6)", but on the other hand, "the fluorescent brightener of formula (1)" of Cited Document 1B invention is "[Formula 2]



(In the formula, a cation corresponding to the sulfonic group is any of cations of an alkali metal and an alkaline earth metal, or an ammonium ion)" (the position at which the sulfonic group is bonded is not specified).

(The different feature 4)

The point in which the "sizing composition" of the Invention includes "(a) at least one binder;

(b) at least one divalent metal salt being selected from the group consisting of calcium chloride, magnesium chloride, calcium bromide, magnesium bromide, calcium iodide, magnesium iodide, calcium nitrate, magnesium nitrate, calcium formate, magnesium formate, calcium acetate, magnesium acetate, calcium sulphate, magnesium sulphate, calcium thiosulphate and magnesium thiosulphate, or mixtures of said compounds; and

(c) water", but on the other hand, Cited Document 1B invention is not specified by this composition.

### (3) Judgment

The judgment regarding the different features 3 and 4 are described as follows.

#### A. The different feature 3

In paragraph [0024] of Cited Document 1, it is described that "aniline 2,5-disulfonic acid" is used as a starting substance, and "a compound represented by formula (1) was obtained"; and the position at which the sulfonic group of formula (1) is bonded in this case becomes identical with that of [formula 1] of the Invention. In addition, the production example is not disclosed in Cited Document 1, in which "aniline disulfonic acid" (example: "aniline 2,4-disulfonic acid") other than "aniline 2,5-disulfonic acid" is used as the starting substance. (Incidentally, also in Japanese Patent Application Publication No. S52-41366 that is of a patent family of Belgium patent No. 719065 which is disclosed in paragraph [0007] of Cited Document 1, the "aniline 2,5-disulfonic acid" and the "aniline 2,4-disulfonic acid" are exemplified, and also it is described that the former is preferable (lines 7 to 10 in left column on page 5); in addition, it is described concerning a light-coloring agent using the former that "the agent is approximately colorless", "1% aqueous solution exhibits strong blue fluorescence", and "the product exhibits an extremely highly pure white color"; but on the other hand, it is described concerning a light-coloring agent using the latter only that the agent "can be separated as a yellowish powder when having salted out" (lines 7 to 11 in right column on page 6 and lines 25 to 27 in right column on page 7).

In consideration of such description of Cited Document 1 as in the above, the different feature 3 is not a substantial different feature. It is only a natural matter at least for a person skilled in the art, who has contacted the description of Cited Document 1, to adopt the composition in which the sulfonic group exists at the position of [Formula 1] of the Invention.

#### B. The different feature 4

The Cited Document 1B invention is the "size press coating liquid", and accordingly it is self-evident that the liquid includes "(a) at least one binder" such as PVA and "(c) water".

In addition, the judgment for the "cationic ink fixing agent" is similar to that of the different feature 1.

#### (4) Effect of the Invention

The effect is similar to that of the above-described 2(4).

#### (5) Summary

The Invention is an invention which a person skilled in the art would have easily made based on Cited Document 1B invention and a well-known art.

### 4 Comparison to Cited Document 2A invention and Judgment

#### (1) Comparison

When the Invention and Cited Document 2A invention are compared to each other, the result is described as follows.

##### A. Sizing composition

The "coating liquid for the receptive layer" of Cited Document 2A invention is "a coating liquid for a receptive layer, which is used in overcoating by an external addition method for ink jet archival paper". Here, the "ink jet archival paper" of Cited Document 2A invention corresponds to "substrates for ink jet printing" of the Invention. In addition, the "sizing composition" of the Invention is "applied" onto "the substrate for ink jet printing" (paragraph [0029] of the detailed description of the Invention of the present application), and is a "liquid" in view of the composition. Accordingly, the "coating liquid for the receptive layer" of Cited Document 2A invention and the "sizing composition" of the Invention are common in the point of the "coating liquid". In addition, the "water-borne liquid composition" of Cited Document 2A invention contains the "fluorescent brightener", and accordingly is a substance that optically brightens the "ink jet archival paper", from the technical view point.

Accordingly, the "coating liquid for the receptive layer" of Cited Document 2A invention and the "sizing composition" of the Invention are common in the point of the "coating liquid that optically brightens the substrate for ink jet printing".

##### B. Binder

The "coating liquid for the receptive layer" of Cited Document 2A invention contains the "polyvinyl alcohol", and the "polyvinyl alcohol" corresponds to "(a) at least one binder" of the Invention in consideration of technical knowledge.

##### C. Water

The "coating liquid for the receptive layer" of Cited Document 2A invention "uses 6 parts of the above described water-borne liquid composition", and "has been adjusted so that the concentration (solid content) of the coating liquid becomes 13%"; and accordingly includes "(c) water", similarly to the Invention.

#### D. Optical brightener of Formula (1)

The "compound of formula (2)" of Cited Document 2A invention is the "fluorescent brightener" and "the cation corresponding to the sulfonic group is the cation of sodium", and accordingly satisfies a requirement of "(d) at least one optical brightener of formula (1)" of the Invention.

### (2) Corresponding features and the different features

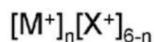
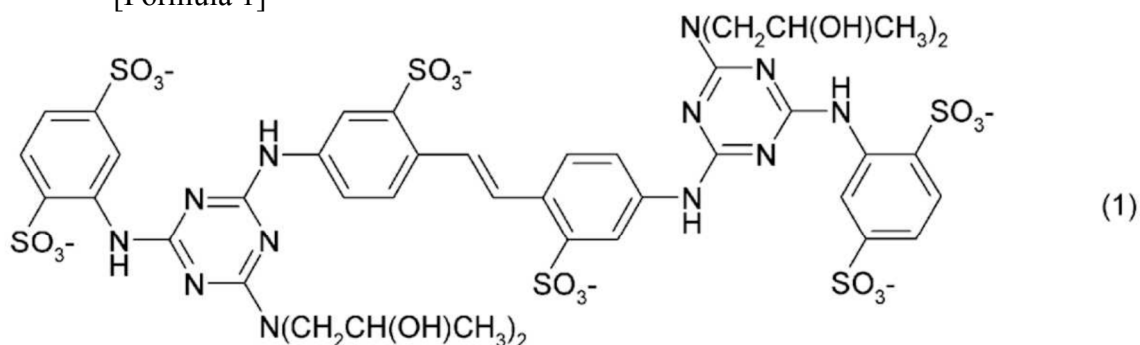
#### A. Corresponding features

The Invention and Cited Document 2A invention correspond with each other in the following constitution.

"A coating liquid for optically brightening a substrate for ink jet printing, including:

- (a) at least one binder;
- (c) water; and
- (d) at least one optical brightener of formula (1).

[Formula 1]



(In the formula, M and X are identical or different, and independently from each other selected from the group consisting of hydrogen, an alkali metal cation, ammonium, ammonium which is mono-, di- or trisubstituted by a C<sub>1</sub>-C<sub>4</sub> linear or branched alkyl radical, ammonium which is mono-, di- or trisubstituted by a C<sub>1</sub>-C<sub>4</sub> linear or branched hydroxyalkyl radical, or mixtures of said compounds, and n is in the range from 0 to 6.)"

#### B. The different features

The Invention and Cited Document 2A invention are different from each other, or are prima facie different from each other, in the following points.

#### (The different feature 5)

The point in which the "coating liquid" of the Invention includes "(b) at least one divalent metal salt being selected from the group consisting of calcium chloride, magnesium chloride, calcium bromide, magnesium bromide, calcium iodide,



magnesium iodide, calcium nitrate, magnesium nitrate, calcium formate, magnesium formate, calcium acetate, magnesium acetate, calcium sulphate, magnesium sulphate, calcium thiosulphate and magnesium thiosulphate, or mixtures of said compounds", but on the other hand, in view of the production step, the "coating liquid for the receptive layer" of the cited invention does not include "(b) at least one divalent metal salt being selected from the group consisting of calcium chloride, magnesium chloride, calcium bromide, magnesium bromide, calcium iodide, magnesium iodide, calcium nitrate, magnesium nitrate, calcium formate, magnesium formate, calcium acetate, magnesium acetate, calcium sulphate, magnesium sulphate, calcium thiosulphate and magnesium thiosulphate, or mixtures of said compounds".

(The different feature 6)

The point in which the "coating liquid" of the Invention is the "sizing composition", but on the other hand, the "coating liquid" of the cited invention is the "coating liquid for the receptive layer".

### (3) Judgment

The judgment regarding the different features 5 and 6 is described as follows.

#### A. The different feature 5

The "coating liquid for the receptive layer" of Cited Document 2A invention includes "polydiallyl dimethyl ammonium chloride (quaternary ammonium salt of polymer)" as the "cationic dye fixing agent "; and a divalent metal salt according to the different feature 3 is well known as a "cationic dye fixing agent" (see the above-described "2" (3) A. (A) to (F).).

In addition to the above-described well-known art, in paragraphs [0005] and [0007] of Cited Document 2, the following are respectively described: "In addition, an anionic dye is used in the ink of an ink jet printer, which uses a water-soluble dye, and accordingly in the archival paper for the ink jet printer of high quality, a cationic ink fixing agent, a water resistant additive, and the like are used in many cases when the archival paper is produced, in order to suppress bleeding of ink on the paper. Furthermore, the archival paper is required to have clear color developability for the image, and accordingly is also required to have a whiteness degree of high quality. Because of this, there are produced sheets of archival paper which are dyed into high whiteness with the use of various fluorescent brighteners. As for the fluorescent brightener that is used at this time, the anionic water-soluble fluorescent brightener is used in many cases from the viewpoint of characteristics in a production process. However, when the above-described cationic fixing agent and the anionic fluorescent brightener are used at the same time, there have been such defects that the ink fixing agent and the fluorescent brightener are combined in the solution to thereby become insoluble in water, and the crystal precipitates, and that such a phenomenon occurs that required high whiteness is not obtained in dyeing." and "[Problem to be solved by the invention] In dyeing of cellulose such as paper, pulp, and cotton with the fluorescent brightener, it has been desired to develop a liquid composition excellent in storage stability at low temperature and high temperature and is also excellent in a brightening effect, and to propose a method for fluorescent brightening the archival paper for an ink jet printer, which is excellent in the fixability of the ink, water resistance, and whiteness degree." As means for solving the problem, such knowledge is disclosed that a

water-borne liquid composition is excellent and contains a fluorescent brightener represented by formula (2) of Cited Document 2A Invention.

In consideration of the above description, it is a matter in a range of usual originality and creativity for a person skilled in the art to independently or concomitantly use the divalent metal salt concerning the different feature 5, as the cationic dye fixing agent of Cited Document 2A Invention.

#### B. The different feature 6

The judgment for the different feature 6 is similar to that for the different feature 2 (above-described 2 (3) B.).

#### (4) Effect of the Invention

The effect of the Invention is similar to that in the above-described 2 (4).

Specifically, in view of the descriptions of [Prior Art] (paragraphs [0002] to [0006]), [Problem to be solved by the invention] (paragraph [0007]), and [Means for solving problem] (paragraphs [0008] to [0013]) of Cited Document 2, the problem and the solving means of the above-described 2(4)A. are only matters that are disclosed also in Cited Document 2.

In addition, in paragraph [0030] of Cited Document 2, it is described that "the water-borne liquid composition of the present invention is excellent in storage stability at low temperature and high temperature under the coexistence of an inorganic salt that has formed in synthesis and in a state of a synthesized sodium salt and the like, without being subjected to steps of removing the inorganic salt, converting the inorganic salt to a lower alkanolamine or a lower alkyl quaternary ammonium salt which enhances solution stability, and adding a solubilizing agent such as urea. In addition, the paper that has been fluorescent-whitened by the water-borne liquid composition of the present invention does not cause a quenching phenomenon; resists being affected by a paper quality and a composition change of the size pressing liquid and the coat liquid; and exhibits excellent whiteness. Furthermore, excellent whiteness degree is obtained even when the water-borne liquid composition of the present invention is used together with a cationic ink fixing agent and a water resistant additive which are usually used in the archival paper for the ink jet printer.", and accordingly effects of the above described 2(4)B. and 2(4)C. are only effects which Cited Document 2A invention shows or are only at least effects which a person skilled in the art expects, who has contacted Cited Document 2.

#### (5) Summary

The Invention is an invention which a person skilled in the art would have easily made based on Cited Document 2A invention and the well-known art.

### 5 Comparison to Cited Document 2B invention and Judgment

#### (1) Comparison

When the Invention and Cited Document 2B invention are compared to each other, the result is as follows.

##### A. Sizing composition

The "size press coating liquid" of Cited Document 2B invention is a "size press coating liquid that provides fluorescent-whitened paper by being applied onto a sheet of

paper with a size pressing machine, and being dried".

Here, the "size press coating liquid" of Cited Document 2B invention corresponds to the "sizing composition" of the Invention. In addition, the "paper" of Cited Document 2B invention and the "substrate for ink jet printing" of the Invention are common in the point of the "substrate." In addition, the "fluorescent-whitened paper" is obtained by the "size press coating liquid" of Cited Document 2B invention, and accordingly the "size press coating liquid" is a liquid that optically brightens the "substrate", from the technical viewpoint.

Accordingly, the "size press coating liquid" of Cited Document 2B invention and the "sizing composition" of the Invention are common in the point of the "sizing composition which optically brightens the substrate".

#### B. Binder

The "size press coating liquid" of Cited Document 2B contains "PVA", and in consideration of technical knowledge, "PVA" corresponds to the "(a) at least one binder" of the Invention.

#### C. Water

The "size press coating liquid" of Cited Document 2B is "prepared from an appropriate mixture containing water and the like", and accordingly includes "(c) water", similarly to the Invention.

#### D. Optical brightener of Formula (1)

The "compound of formula (2)" of Cited Document 2B invention is a "fluorescent brightener", and includes the invention that the "cation corresponding to the sulfonic group" is the "cation of an alkali metal".

Then, the "compound of formula (2)" of Cited Document 2B invention satisfies a requirement of "(d) at least one optical brightener of formula (1)" of the Invention.

### (2) Corresponding features and the different features

A. The Invention and Cited Document 2B invention correspond with each other in the following constitution.

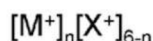
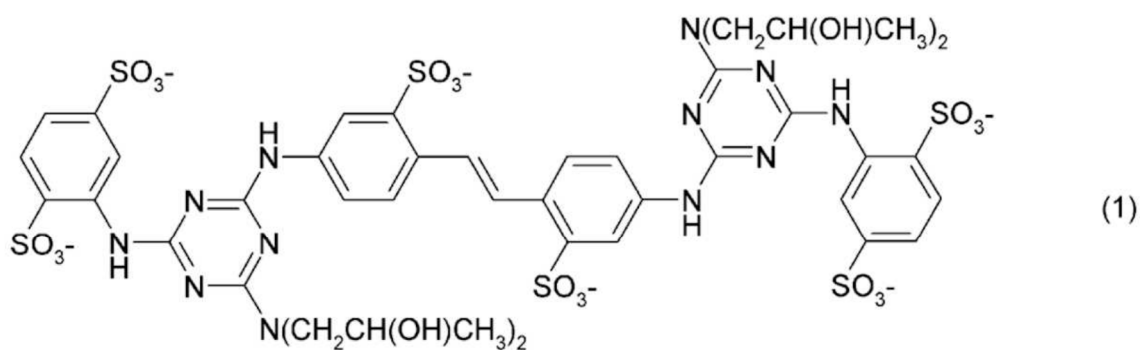
"A sizing composition which optically brightens a substrate, including:

(a) at least one binder;

(c) water; and

(d) at least one optical brightener of formula (1).

[Formula 1]



(In the formula,

M and X are identical or different, and independently from each other selected from the group consisting of hydrogen, an alkali metal cation, ammonium, ammonium which is mono-, di- or trisubstituted by a C<sub>1</sub>-C<sub>4</sub> linear or branched alkyl radical, and ammonium which is mono-, di- or trisubstituted by a C<sub>1</sub>-C<sub>4</sub> linear or branched hydroxyalkyl radical, or mixtures of said compounds, and n is in the range 0 to 6.)"

B. The Invention and Cited Document 2B Invention are different from each other in the following points.

(The different feature 7)

The point in which the "substrate" of the Invention is the "substrate for ink jet printing", and the "sizing composition" of the Invention includes "(b) at least one divalent metal salt being selected from the group consisting of calcium chloride, magnesium chloride, calcium bromide, magnesium bromide, calcium iodide, magnesium iodide, calcium nitrate, magnesium nitrate, calcium formate, magnesium formate, calcium acetate, magnesium acetate, calcium sulphate, magnesium sulphate, calcium thiosulphate and magnesium thiosulphate, or mixtures of said compounds", but on the other hand, the "substrate" of Cited Document 2B invention is "paper" that is not specified as paper "for ink jet printing", and also is not defined so as to include "(b) at least one divalent metal salt being selected from the group consisting of calcium chloride, magnesium chloride, calcium bromide, magnesium bromide, calcium iodide, magnesium iodide, calcium nitrate, magnesium nitrate, calcium formate, magnesium formate, calcium acetate, magnesium acetate, calcium sulphate, magnesium sulphate, calcium thiosulphate and magnesium thiosulphate, or mixtures of said compounds".

### (3) Judgment

The judgment regarding the different feature 7 is as follows.

The "paper" of Cited Document 2B invention is the "fluorescent-whitened paper", and accordingly is suitable for "ink jet printing", and the "paper" of Cited Document 2B invention is "paper" which is obtained by size press by an external addition method, and accordingly it is a matter of naturally being carried out by a person skilled in the art to prevent the ink of ink jet printing from bleeding by making the size press coating liquid contain a cationic ink fixing agent. In addition, the divalent metal salt concerning the different feature 6 is well known as the cationic ink fixing agent (see the above-described "2"(3)A. (A) to (F)).

Then, it is a matter which a person skilled in the art would have easily achieved when developing the application of the "paper" of Cited Document 2B invention, to adopt the constitution regarding the different point 7 in Cited Document 2B invention.

(4) Effect of the Invention

The effect is similar to that of the above-described 4(4).

(5) Summary

The Invention is an invention which a person skilled in the art would have easily made, based on Cited Document 2B invention and the widely well-known art.

6 Appellant's allegation

The appellant alleges that "the cationic ink fixing agent described in Cited Documents 1 and 2 is, actually, an organic compound. On the other hand, the divalent metal salt that is used in the Invention is an inorganic compound. In Cited Documents 1 and 2, it is described that "usable cationic ink fixing agents include: higher fatty amine, a quaternary-ammonium-salt type compound, an ethylene oxide addition product of a secondary alkylamine, a cationic polymer compound, inorganic particles whose surfaces are cationically charged, and the like. ("paragraph [0018] of Cited Document 1, and paragraph [0027] of Cited Document 2)".

However, the divalent metal salt such as calcium chloride is well known as the cationic ink fixing agent, and accordingly, it is clear for a person skilled in the art that the divalent metal salt such as the calcium chloride is included in "the like" of "Usable cationic ink fixing agents include: higher fatty amine, a quaternary-ammonium-salt type compound, an ethylene oxide addition product of a secondary alkylamine, a cationic polymer compound, inorganic particles whose surfaces are cationically charged, and the like." (paragraph [0018] in Cited Document 1, and paragraph [0027] in Cited Document 2).

Incidentally, a reaction liquid of the fluorescent brightener described in Cited Document 1 and Cited Document 2 could not help but contain a large amount of chlorine that is derived from cyanuric chloride in the production process (where the reaction liquid results in containing 6 moles of chlorine in terms of sodium chloride with respect to 1 mole of the fluorescent brightener.). In addition, a cation corresponding to the sulfonic group of the fluorescent brightener described in Cited Document 1 and Cited Document 2 may be an alkaline earth metal; specifically, calcium or magnesium. Then, it is a matter which a person skilled in the art, who had contacted Cited Document 1 and Cited Document 2, would have naturally understood that the fluorescent brightener is highly stable even though the divalent metal salt such as calcium chloride is contained in the solution of the fluorescent brightener described in Cited Document 1 and Cited Document 2. In addition, it is also disclosed in paragraphs [0008] and [0009] of Cited Document 1 (paragraphs [0019] and [0020]) of Cited Document 2) that an aqueous solution of a fluorescent brightener is obtained by dissolving crystals of the fluorescent brightener in water, which have been salted out by an alkali metal salt or an alkaline earth metal salt, (that the crystals dissolve only by being mixed with water), and that the obtained reaction liquid may be condensed and used (that the obtained reaction liquid is highly stable even when having been condensed).

When the above-described circumstance is taken into consideration, it can be said that even though the divalent metal salt such as the calcium chloride is not explicitly shown in paragraph [0018] of Cited Document 1 and paragraph [0027] of Cited Document 2, the descriptions in the above paragraphs are considered to practically exemplify the divalent metal salt such as the calcium chloride as the cationic ink fixing agent, for a person skilled in the art, who has contacted the Cited Document 1 and the Cited Document 2.

### No. 3 Summary

The Invention would have been easily made by a person who had ordinary skill in the art belonging to the Invention before the priority date, based on the invention described in Cited Document 1 or Cited Document 2 which had been distributed in Japan or a foreign country before the priority date of the present application, and on the well-known art, and accordingly, the patent should not be granted under the provisions of Article 29 (2) of the Patent Act.

Accordingly, the present application should be rejected without examining other claims.

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

June 6, 2016

Chief administrative judge: TETSU, Toyoo  
Administrative judge: HIGUCHI, Nobuhiro  
Administrative judge: SHIMIZU, Yasushi