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(54) HEAT-SUBLIMABLE INK RIBBON.

(57) A heat-sublimable ink ribbon comprising a heat-resistant base sheet and a heat-sublimable ink formed on the base sheet surface. The ink comprises a sublimable ink, a binder, and 30 to 200 parts by weight, per 100 parts by weight of the sum of the dye and the binder, of solvent-insoluble particles with high thermal conductivity uniformly dispersed in the binder and dye. The use of this ribbon enables the dye to be transferred through sublimation effectively by slight heat and prevents welding between the ribbon and print paper by heat.

Croydon Printing Company Ltd

DESCRIPTION

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TECHNICAL FIELD

The present invention relates to a dye carrier ink ribbon for a sublimation transfer type hard copy, which is used to make a color picture image on a printing paper by thermally transferring the dye selectively from the carrier ink ribbon to the printing paper.

BACKGROUND ART

A color copying method has hitherto been proposed that a dye carrier ink ribbon in which an ink formed by dissolving and dispersing a sublimable dye into resin and solvent is coated on a thin heat resisting sheet such as paper or the like is used and which is heated from its back side by a thermal print head to thereby sublimate only the dye contained in the ink, so that the dye is transferred on a printing paper coated on its surface with a resin of excellent dye absorbing property such as polyester resin to form a color picture image.

The dye carrier ink ribbon used in this color copying method was requested to be capable of efficiently sublimating the dye on the printing paper so that the coloring concentration of picture image is increased to obtain a clear picture image of high contrast and that heat energy applied to the thermal print head of printer is reduced to decrease power consumption and the burden of thermal print head, the life of thermal print head being lengthened.

Moreover, the dye carrier ink ribbon used in this color copying method is requested to be capable of efficiently sublimating the dye and to be prevented from being melt bonded by heat on a coating composition coated on the printing paper.

Generally in such color copying method, since the dye contained in the dye carrier ink ribbon is instantaneously sublimated by the thermal print head heated at a high temperature of 200°C or above and transferred on the printing paper which contacts with the dye carrier ink ribbon, a melt bonding phenomenon easily occurs in which the binder resin and dye contained in the ink and the coating composition coated on the printing paper are melted by heat and adhered to each other. When the above melt bonding occurs, the ink ribbon becomes difficult to be released from the printing paper. Particularly in the portion having high color concentration, the base material is damaged. In addition, the melting phenomenon occurs part by part, and particularly in the intermediate gradation of the picture image, such melt bonding phenomenon appears as roughness of picture image. In the case of mixed colors, particularly the concentration of only the portion in which the melted portions are superposed on each other is increased to thereby lower picture quality. As a result, the clear intermediate gradation of the picture could not be obtained. To solve these problems, it has been proposed that the resin itself contained in the ink is substituted with a resin of enoughly high heat resisting property or that the resin is cured by curing agent or like means to increase the heat resisting property and the heat resisting property of the printing paper coating composition is similarly increased whereby to prevent the resins from being melted with each other. However, with the above methods, the resin and dye in the above intermediate gradation can not sufficiently be prevented from being melt bonded with each other. While, it is general that as the heat resisting property of resin is increased, the resin covering the dye is difficult to be softened by heat generated from the thermal print head upon printing and then probability of the occurrence of the melt bonding phenomenon is reduced, however the amount of dye transferred on the printing paper is decreased and thus the coloring concentration is apt to be lowered.

DISCLOSURE OF INVENTION

The present invention relates to a dye carrier ink ribbon for a sublimation transfer type hard copy formed of a base sheet having heat resisting property and an ink containing sublimable dye formed on the surface of the base sheet. The ink is formed of a sublimable dye, a binder and 30 to 200 parts by weight of powder of high thermal conductivity insoluble to a solvent uniformly dispersed into totally 100 parts by weight of the sublimable dye and the binder. According to the present invention, the sublimable dye can efficiently be sublimated and transferred on the printing paper by heat and the melt bonding between the dye carrier ink ribbon and the printing paper is avoided.

BEST MODE FOR CARRYING OUT THE INVENTION

An ink used in a color copying method of sublimation transfer type is generally prepared by mixing a sublimable dye and a binder resin containing in a curable resin and a solvent added with small amount of a dispersion agent and a lubricant. If necessary, for the purpose of improving the printing property and of adjusting viscosity, about 10 parts by weight of inorganic fine particle is sometimes added to the ink binder.

The present inventor has found out that the dye carrier ink ribbon formed by coating on a base sheet having heat resisting property such as a paper or the like an ink in which as much as 30 to 200 parts by weight of inorganic powder having excellent thermal conductivity, and insoluble to a solvent of the ink are dispersed for 100 parts by weight of the sublimable dye and the binder is considerably effective for particularly the color copying method.. The experimental results proving such effect will be described hereinafter.

First, an ink was formed with a mixing 4 parts by weight of SUMIKAPLAST BLUE S35 (manufactured by Sumitomo Chemical Co., Ltd.) as a sublimable dye, 6 parts by weight of acetate cellulose (manufactured by Daicel Ltd.) as a binder and 90 parts by weight of methyl ethyl ketone as a solvent. And, experiments 1 to 8 were carried out in which silica (AEROSIL R972, manufactured by Nippon Aerosil Co., Ltd.) was mixed into the above ink with various mixing ratios within the upper limit of 250 parts by weight relative to totally 100 parts by weight of the sublimable dye and the binder. In the respective experiments, the mixed compositions were dispersed by a ball mill for one day and coated on a condenser paper of 25 μ thickness to have a coating amount of 3 g/m^2 after being dried thus dye carrier ink ribbons being formed. After the ink surface of the dye carrier ink ribbon was made in contact with the surface treated printing paper in which the surface is coated by polyester resin, the dye carrier ink ribbon was heated at its back side by controlling the current of the thermal print head to gradually lower the temperature from 300°C at maximum so that the sublimated amount of dye was changed and at the same time, the printing paper and the dye carrier ink ribbon were moved to thereby form a gradation picture image. Then, the melt bonded condition between the ink robbon and the printing paper and the finished condition of the intermediate gradation after the printing were observed. Also, the coloring concentration of cyan at the highest concentration portion of the gradation picture image formed on the printing paper was measured by a Macbeth reflection density meter. The experimental results of experiment 1 in which silica concentration was taken as 0 parts by weight (hereinafter simply described as "silica concentration 0 parts by weight") relative to 100 parts by weight of sublimable dye and acetate cellulose binder, of the experiment 2 in which the silica concentration was taken as 20 parts by weight, of the experiment 3 in which the silica concentration was taken as 30 parts by

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weight, of the experiment 4 in which the silica concentration was taken as 50 parts by weight, of the experiment 5 in which the silica concentration was taken as 100 parts by weight, of the experiment 6 in which the silica concentration was taken as 150 parts by weight and of the experiment 7 in which the silica concentration was taken as 200 parts by weight and of the experiment 8 in which the silica concentration was taken as 250 parts by weight will hereinafter be described with reference to a table 1.

In the experiment-1 in which the silica concentration was selected to be 0 parts by weight, the dye carrier ink ribbon and the printing paper after printing were completely melt bonded with each other (hereinafter, simply described as "melt bonding"). The melt bonding and roughened finish were frequently found out in the finished intermediate gradation of the coloring concentration. And, it was impossible to measure the cyan coloring concentration (hereinafter, simply described as coloring concentration) of the highest concentration portion of the gradation picture image formed on the printing paper.

In the experiment-2 in which the silica concentration was selected to be 20 parts by weight, the gradation picture image was melt bonded at its high concentration portion, the finished intermediate gradation was frequently roughened and the coloring concentration was 1.31.

In the experiment-3 in which the silica concentration was selected to be 30 parts by weight, no melt bonding occurred, the finished intermediate gradation was satisfactory and the coloring concentration was 1.65.

TABLE-1

experiment No.	silica concentration (parts by weight)	coloring concentration	finished intermediate gradation	melt bonding
experiment-1	0	unmeasurable	frequently melt bonded and roughened frequently	completely melt bonded
experiment-2	20	1.31	frequently roughened	melt bonded at high concentration portion
experiment-3	30	1.65	satisfactory	not melt bonded
experiment-4	50	1.72	satisfactory	not melt bonded
experiment-5	100	1.68	satisfactory	not melt bonded
experiment-6	150	1.65	satisfactory	not melt bonded
experiment-7	200	1.57	satisfactory	not melt bonded
experiment-8	250	1.03	powders partially came off	not melt bonded

In the experiment-4 in which the silica concentration was selected to be 50 parts by weight, no melt bonding occurred, the finished intermediate gradation was satisfactory and the coloring concentration was 1.72.

In the experiment-5 in which the silica concentration was selected to be 100 parts by weight, no melt bonding occurred, the finished intermediate gradation was satisfactory and the coloring concentration was 1.68.

In the experiment-6 in which the silica concentration was selected to be 150 parts by weight, no melt bonding occurred, the finished intermediate gradation was satisfactory and the coloring concentration was 1.65.

In the experiment-7 in which the silica concentration was selected to be 200 parts by weight, no melt bonding occurred, the finished intermediate gradation was satisfactory and the coloring concentration was 1.57.

In the experiment-8 in which the silica concentration was selected to be 250 parts by weight, although no melt bonding occurred, the finished intermediate gradation was poor, powders partially came off and the coloring concentration was 1.03.

Further, experiments-9 to -14 in which the mixed particle was not limited to silica but heat conductive powder particles were added were carried out by the present inventor. These experiments were carried out as follows.

50 parts by weight of various powder particles having excellent heat conductivity were added to 100 parts by weight of solid component of the ink formed with a mixing ratio of 13 parts by weight of a sublimable dye, KAYASET RED 126 (manufactured by Nippon Kayaku Co., Ltd.), 10 parts by weight of ethyl cellulose, 10 parts by weight of melamine resin, 0.2 parts by weight of paratoluen sulfonic acid and 133 parts by weight of methyl ethyl ketone. This product was dispersed for 5 hours by a sand

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mill treatment and thus an ink was formed. This ink was then coated on a rice paper of area weight of 20 g/m^2 so as to have a coating weight of 3 g/m^2 after being dried and thereafter heated for 3 minutes at 80°C and for one hour at 100°C to harden the binder resin thus a dye carrier ink ribbon was formed. And, the experiments-9 to -14 in which the powder particle of excellent heat conductivity to be added was changed and the mixing ratio thereof was changed were carried out and the experimental results indicated on table 2 were obtained. The same terms as those on the table-1 were used for the coloring concentration, the finished intermediate gradation and the melt bonding.

In the experiment-9 in which no powder particle was added at all, the high concentration portion was melt bonded, the finished intermediate gradation was roughened and the coloring concentration was 1.30.

TABLE-2

experiment No.	added powder particle & its mixed ratio	coloring concentration	finished intermediate gradation	melt bonding
experiment-9	none	1.30	frequently roughened	melt bonded at high concentration portion
experiment-10	silica powder 100%	1.55	satisfactory	not melt bonded
experiment-11	silica powder 70% carbon powder 30%	1.68	satisfactory	not melt bonded

Cont'd

experiment No.	added powder particle & its mixed ratio	coloring concentration	finished intermediate gradation	melt bonding
experiment-12	silica powder 70% aluminium powder 30%	1.73	satisfactory	not melt bonded
experiment-13	silicone carbide 100%	1.70	satisfactory	not melt bonded
experiment-14	silica powder 50% calcium carbonate 50%	1.56	satisfactory	not melt bonded

In the experiment-10 in which 50 parts by weight of silica powder was only added, no melt bonding occurred, the intermediate gradation was finished satisfactorily and the coloring concentration was 1.55.

In the experiment-11 in which 50 parts by weight of mixture of 70% of silica powder and 30% of carbon powder was added, no melt bonding occurred, the intermediate gradation was finished satisfactorily and the coloring concentration was 1.68.

In the experiment-12 in which 50 parts by weight of mixture of 70% of silica powder and 30% of aluminium powder was added, no melt bonding occurred, the intermediate gradation was finished satisfactorily and the coloring concentration was 1.73.

In the experiment-13 in which only 50 parts by weight of silicone carbide powder was added, no melt bonding occurred, the intermediate gradation was finished satisfactorily and the coloring concentration was 1.70.

In the experiment-14 in which totally 50 parts by weight of silica powder and calcium carbonate with a mixed ratio of 50% and 50% was added, no melt bonding occurred, the intermediate gradation was finished satisfactorily and the coloring concentration was 1.56.

Heat generated by the thermal print head is conducted through the sheet having heat resisting property, which forms the base of the ink ribbon, to the ink layer to heat the dye to its sublimation starting temperature whereby a picture image is formed by sublimating and transferring the dye from the ink ribbon to the surface of the printing paper. In this case, the heat generated by the thermal print head must instantaneously be conducted to the ink layer effectively. From these experiments, it was understood that when the thermal conductive powder particles were dispersed with high concentration in the ink, the heat conducted through the heat resisting sheet was effectively conducted into the ink to rapidly start the sublimation of the sublimable dye contained in the ink. The effect of the present invention can not be achieved by only increasing the thermal conductivity of the ink ribbon. For example, when a metal foil is used or metallization is carried out so as to increase the thermal conductivity of the heat resisting property sheet, the heat of the thermal print head is increasingly conducted in the lateral direction so that the ink is not heated effectively thus the sublimation property of the dye being lowered. On the other hand, the powder particles dispersed with high concentration in the ink of the present invention partially contact with each other, in which when the particle is dominantly heated, the dye dispersed and adsorbed in the periphery of the particle can effectively be heated and sublimated. However, since the thickness of the ink coating film and the contact area are small, the conductions of heat in the lateral and thickness directions were not so different from each other so that the ink ribbon of excellent sublimation property and of high resolution could be obtained. The present invention is more effective if the coating surface of the ink ribbon according to the experimental example of the present invention is made concave and convex in a quite small area by highly packed powder particle with the result that the surface area of the ribbon is increased. Accordingly, since a space in which the dye heated by the conducted heat can be sublimated was increased, the amount of dye transferred on the surface of the printing paper was increased and thus the picture image of excellent coloring property was obtained.

On the other hand, since the concave and convex portions formed on the surface of the ink ribbon of the present experimental examples lower the contact area with the printing paper, there is then an effect that the ink ribbon and the printing paper can be prevented from being melt bonded by heat with each other. Particularly, the heat of the thermal print head was effectively conducted by the powder particle of excellent thermal conductivity packed with high concentration into the ink and the sublimation space was increased by the concave and the convex portions formed on the surface of the ink ribbon to increase the sublimation efficiency so that even when a heat resisting property resin of poor sublimation efficiency for the prior art ribbon was used, a picture image of enoughly high coloring concentration could be formed on the printing paper. As a result, it was particularly effective that the melt bonding on the high concentration portion of the formed picture image was removed and further the minute melt bonding in the intermediate gradation area could be avoided whereby a clear image having no scattered dye concentration could be formed over a range from the high concentration area through the intermediate gradation area to the low concentration area. From the results of the experiments-1 to -8, it could be confirmed that the above effect could be achieved by dispersing into totally 100 parts by weight of the ink binder containing the sublimable dye, 30 to 200, preferably 40 to 150 parts by weight of powder particle of excellent thermal conductivity insoluble to the solvent. When the adding amount is less than 30 parts by weight, the particles do not contact with one another sufficiently and the surface of the ink ribbon is not formed to have quite small concave and convex portions so that the heat can not be conducted effectively. Also, since the sublimation space is small, the sublimation efficiency can not be increased sufficiently. In addition, the melt bonding phenomenon between the ink ribbon and the printing paper is frequently caused and the melt bonding occurred particularly on the intermediate gradation area can not be avoided. When the adding amount exceeds 200 parts by weight, the powder particle is transferred to the side of the printing paper upon printing, and the dye concentration is reduced too much and thus the coloring concentration becomes insufficient. In this case, the diameter of the powder particle is preferably selected to be less than 100 μ, more preferably in a range from 10 mμ to 10 μ.

In order to get a knowledge of necessary adding ratio of the powder particle, the following experiments-15 to -17 were carried out.

Resin with a mixed ratio of 10 parts by weight of ethyl cellulose, 10 parts by weight of rapid-curing type melamine resin and 0.2 parts by weight of paratoluene sulfonic acid was mixed with a sublimable dye PS Blue RR (manufactured by Mitsui Toatsu Chemicals, Inc.) so as to satisfy the mixing ratios of the binder to the sublimable dye being 3:1, 3:2 and 3:3. Then, the mixture was diluted by methyl ethyl ketone to 10%.

Powder particles of excellent thermal conductivity in which silica powder (AEROSIL R972, manufactured by Nippon Aerosil Co., Ltd.) and silicone carbide powder (manufactured by Fujimi Kenmazai Kogyo Kabushiki Kaisha) were mixed to each other with a weight ratio of 3:2 were added to 100 parts by weight of the ink containing the binder and the dye, which then was dispersed for 15 hours by an attrition mill, thus an ink was formed. The above ink was coated on a condenser paper of area weight of 20 g/m^2 in such a manner that the thickness of the ink coating was changed so as to make the dyes in the constant area substantially same between the ink ribbons containing different amount of powder after being dried. Then ink ribbons were formed and heated at 80°C for 15 hours and the resin was cured. After that, similarly to the experiment-1, the prints were made on the printing paper by using the ink ribbon, and the cyan concentrations of the picture images at its highest concentration portion were measured and compared. In this case, the measured value represents the cyan concentration measured by the Macbeth reflection density meter.

The experiment-15, the experiment-16 and the experiment-17 will be described with reference to a table-3.

In the experiment-15 in which the mixing ratio of the sublimable dye and the resin was selected to be 1:3, when the adding amount of the powder particle relative to 100 parts by weight of the sublimable dye and the resin was selected to be 0 parts by weight, the cyan concentration at the highest concentration portion was 0.98, when it was selected to be 30 parts by weight, the cyan concentration at the highest concentration portion was 1.48 and when it was selected to be 70 parts by weight, the cyan concentration at the highest concentration portion was 1.50.

In the experiment-16 in which the mixing ratio of the sublimable dye and the resin was selected to be 2:3, when the adding amount of the powder particle relative to 100 parts by weight of the sublimable dye and the resin was selected to be 0 parts by weight, the cyan concentration at the highest concentration portion was 1.23, when it was selected to be 30 parts by weight, the cyan concentration at the highest concentration portion was

1.60 and when it was selected to be 70 parts by weight, the cyan concentration at the highest concentration portion was 1.65.

TABLE-3

experiment	(dye):(resin)	the adding ratio of powder particle		
		0 parts by weight	30 parts by weight	70 parts by weight
experiment-15	1 : 3	0.98	1.48	1.50
experiment-16	2 : 3	1.23	1.60	1.65
experiment-17	3 : 3	1.35	1.72	1.76

In the experiment-17 in which the mixing ratio of the sublimable dye and the resin was selected to be 3:3, when the adding amount of the powder particle relative to 100 parts by weight of the sublimable dye and the resin was selected to be 0 parts by weight, the cyan concentration at the highest concentration portion was 1.35, when it was selected to be 30 parts by weight, the cyan concentration at the highest concentration portion was 1.72 and when it was selected to be 70 parts by weight, the cyan concentration at the highest concentration portion was 1.76.

From the above three experiments, it was understood that regardless of the mixing ratio of the sublimable dye and the resin, if about 30 parts by weight of powder particle of excellent thermal conductivity was mixed into 100 parts by weight of sublimable dye and resin, the satisfactory coloring concentration of the picture image could be obtained.

As will be clear from the various experimental results as described above, according to the dye carrier ink ribbon formed by coating on the surface of the base sheet having heat resisting property the ink in which 30 to 200 parts by weight of particle of high thermal conductivity and insoluble to the solvent were dispersed into totally 100 parts by weight of the ink formed of the sublimable dye and the binder, particularly the sublimable dye can efficiently be sublimated and transferred on the printing paper by a small amount of heat and it is possible to prevent the ink ribbon and the printing paper from being melt bonded with each other by heat. The powder particle of excellent thermal conductivity and insoluble to the solvent which is used in the present invention is not limited to inorganic powder particle such as titanium oxide, zinc oxide, calcium carbonate, barium sulfate, aluminium oxide, silica, clay, magnesium oxide, tin oxide, silicone carbide, beryllia, glass powder and the like but may be metal powder such as iron powder, copper powder, aluminium powder and the like and carbide such as graphite, carbon and the like. Since these powder particles have high thermal conductivity as compared with the heat resisting plastic sheet or paper which becomes the base material of the ink ribbon, and the resin contained as the binder, the heat conducted from the thermal print head can effectively be conducted into the ink. In general, if mainly inexpensive inorganic powder particle such as calcium carbonate, clay, silica or the like, if necessary, metal powder such as iron powder, aluminium powder and the like and carbon are mixed into the ink, the effect of the present invention becomes greater. In addition, short fibre-like and phosphor piece-like powders may be added to the ink with a small amount which does not lower the printing property.

CLAIMS

1. A dye carrier ink ribbon for a sublimation transfer type hard copy comprising a base sheet having heat resisting property and a thermally sublimable ink formed on the surface of said base sheet, characterized in that said dye carrier ink is made of a sublimable dye, a binder and 30 to 200 parts by weight of powder particle of high thermal conductivity and insoluble to solvent dispersed into totally 100 parts by weight of said sublimable dye and said binder.

2. A dye carrier ink ribbon used for a sublimation transfer type hard copy in which under the condition that an ink ribbon for supporting a sublimable dye and a printing paper in contact with each other, a picture image is formed on the surface of said printing paper by selectively heating said sublimable dye contained in said ink ribbon, comprising a base sheet having heat resisting property and an ink formed on the surface of said base sheet, characterized in that said dye carrier ink is made of a sublimable dye, a binder and 30 to 200 parts by weight of powder particle of high thermal conductivity and insoluble to solvent dispersed into totally 100 parts by weight of said sublimable dye and said binder.

3. A dye carrier ink ribbon according to claim 1 or 2, characterized in that the adding amount of said powder particle is selected in a range from 40 to 150 parts by weight.

4. A dye carrier ink ribbon according to claim 1, 2 or 3, characterized in that said powder particle is selected from titanium oxide, zinc oxide, calcium carbonate, barium sulfate, aluminium oxide, clay, silica, magnesium oxide, tin oxide, silicone carbide, beryllia, glass powder, metal particle, graphite and carbon.

INTERNATIONAL SEARCH REPORT

International Application No. PCT/JP83/00375

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) [8]

According to International Patent Classification (IPC) or to both National Classification and IPC

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II. FIELDS SEARCHED

Minimum Documentation Searched [4]

Classification System	Classification Symbols
I P C	B41M 5/26

Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched [5]

Kokai Jitsuyo Shinan Koho 1971 - 1983

III. DOCUMENTS CONSIDERED TO BE RELEVANT [14]

Category*	Citation of Document, [16] with indication, where appropriate, of the relevant passages [17]	Relevant to Claim No. [18]
X	JP,A, 56-109787 (Fuji Kagakushi Kogyo Kabushiki Kaisha, Toshiba Corp.), 31. August. 1981 (31. 08. 81)	1 - 4
X	JP,A, 52-99109 (Reiko Kabushiki Kaisha), 19. August. 1977 (19. 08. 77)	1 - 4
X	JP,A, 48-79017 (Dainippon Printing Co., Ltd.), 23. October. 1973 (23. 10. 73), P. 5	1 - 4

* Special categories of cited documents: [15]

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

IV. CERTIFICATION

Date of the Actual Completion of the International Search [2]	Date of Mailing of this International Search Report [2]
January 5, 1984 (05. 01. 84)	January 17, 1984 (17. 01. 84)
International Searching Authority [1]	Signature of Authorized Officer [20]
Japanese Patent Office	