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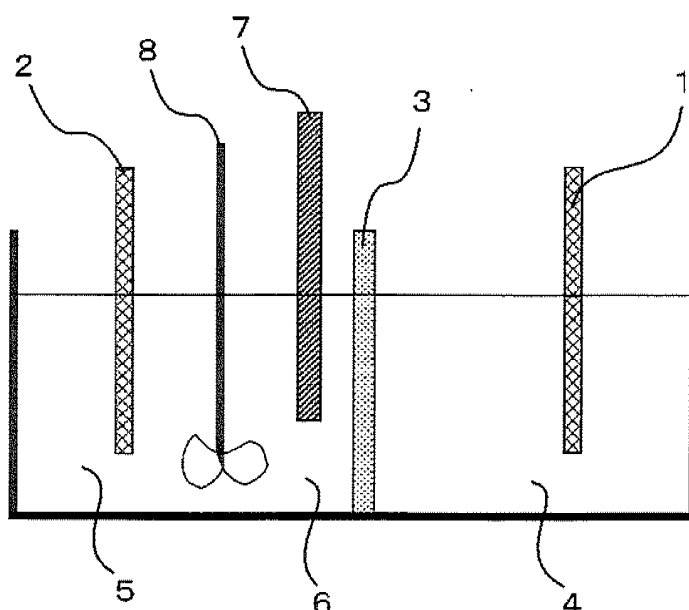
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(54) **METHOD AND APPARATUS FOR DEHALOGENATING ORGANIC HALIDE THROUGH ELECTROLYSIS**

(57) An object is to provide a method for completely and fully dehalogenating an organic halide through electrolysis, and an electrolysis apparatus used therefor. One embodiment of the invention is a method wherein, when carrying out dehalogenation by electrolyzing an organic halide or a liquid containing the organic halide, a diaphragm electrolytic cell in which an anode and a cathode are separated is used, and electrolysis is carried out while

applying ultrasonic waves using ultrasonic wave generating means and stirring using stirring means, and an electrolytic cell used therefor. Another embodiment of the present invention is a method wherein, when carrying out electrolysis, a hydrogen-absorbing metal or alloy is used as a cathode. In this case also, it is preferable to further use a diaphragm electrolytic cell, and carry out electrolysis using ultrasonic wave generating means and stirring means.

FIG. 1



Description

[0001] The present invention relates to a method for dehalogenating an organic halide through electrolysis so as to render it harmless, and apparatus used therefor.

[0002] Organic halides such as polychlorinated biphenyls (PCBs) or dioxins are generally harmful to the human body, and many thereof pollute the environment. In particular, PCBs cause a serious social problem as an environmental pollutant since they are resistant to degradation, remain for a long period of time without decomposing, and continue to pollute the surroundings. Furthermore, PCBs also have the problem that it is difficult to process them so as to render them harmless when they are disposed of, etc. since they are chemically very stable substances.

[0003] With regard to methods that are currently allowed by the Waste Disposal Law regulating the disposal of waste oil, etc. containing PCBs, there are known a high temperature thermal decomposition method, a dechlorinating decomposition method, a hydrothermal oxidative decomposition method, a reductive thermochemical decomposition method, a photodecomposition method, and a plasma decomposition method. Among these, from the viewpoint of especially large-sized and specialist decomposition equipment being unnecessary, no harmful products being formed, the possibility of reuse of by-products, etc. the dechlorinating decomposition method is preferable.

[0004] In general, in a dehalogenating decomposition method, by mixing sodium metal, an organic alkali metal, a catalyst, etc. with an organic halide and carrying out a chemical reaction, the halogen in the organic halide is replaced with hydrogen, etc. (ref. e.g. Patent Publications 1 to 3 below). However, there are the defects and problems that the metal and catalyst used in the dehalogenating decomposition are expensive and it is necessary to carry out this chemical reaction in an anhydrous organic solvent.

[0005] As a method for making PCBs, etc. harmless by a relatively simple procedure and means, there has been proposed a method in which reduction/dehalogenation is carried out by adding and mixing calcium metal with an alcoholic solution of a PCB, etc. (ref. Patent Publication 4 below). However, this method also has the problem that it is necessary to use an organic solvent.

[0006] The present inventors have also proposed a method in which a PCB and a solvent such as water or acetonitrile are mixed, and the PCB is treated by electrolysis (ref. Patent Publication 5 below). This method is an excellent method since the PCB, etc. can be made harmless relatively simply. However, in order to carry out dehalogenation completely by this method; it is necessary to modify the conditions and equipment for the electrolysis.

(Patent Publication 1) JP-A-2002-756 (JP-A denotes a Japanese unexamined patent application publication)

(Patent Publication 2) JP-A-2001-269673

(Patent Publication 3) JP-A-8-66494

(Patent Publication 4) JP-A-2002-265391

(Patent Publication 5) JP-A-2002-345991

[0007] It is an object of the present invention to provide a method for completely and fully dehalogenating an organic halide through electrolysis, and an electrolysis apparatus used therefor.

[0008] One embodiment of the present invention is a method for dehalogenating an organic halide, wherein when carrying out dehalogenation by electrolyzing the organic halide or a liquid containing the organic halide, a diaphragm electrolytic cell in which an anode and a cathode are separated is used, and electrolysis is carried out while applying ultrasonic waves using ultrasonic wave generating means and stirring using stirring means.

[0009] Another embodiment of the present invention is a method for dehalogenating an organic halide, wherein when carrying out dehalogenation by electrolyzing the organic halide or a liquid containing the organic halide, electrolysis is carried out using as a cathode a hydrogen-absorbing metal or alloy.

[0010] Yet another embodiment of the present invention is an electrolytic cell for carrying out dehalogenation by electrolyzing an organic halide or a liquid containing the organic halide, wherein the electrolytic cell comprises an anode and a cathode separated by a diaphragm, ultrasonic wave generating means, and stirring means.

[0011] In accordance with the present invention, since a toxic organic halide such as a PCB or a dioxin can be treated at normal temperature and normal pressure, the treatment operation is safe. Furthermore, since these organic halides are decomposed using electricity and water so as to make them harmless, the means and method are relatively simple, inexpensive, and clean in terms of the reaction environment. In the present invention, due to the emulsion effect of ultrasonic waves generated by the ultrasonic wave generating means, micro clusters are formed from the organic halide or from the organic halide and, for example, water. Due to the cavitation effect of ultrasonic waves, an environment with a high pressure of 1000 atm or higher and a high temperature of 5000°C or higher is formed at the moment a bubble in the cluster bursts. In the present invention, due the emulsion effect and the cavitation effect, dehalogenation through electrolysis is carried out very efficiently.

[0012] (FIG. 1) A schematic diagram of an electrolytic cell of the present invention.

- 1 Anode
- 2 Cathode
- 3 Diaphragm
- 4 Anode chamber
- 5 Cathode chamber
- 6 Liquid to be treated
- 7 Ultrasonic wave oscillator rod
- 8 Stirring means

[0013] The present invention is characterized in that,

when carrying out dehalogenation by electrolyzing an organic halide or a liquid containing the organic halide, a diaphragm electrolytic cell in which an anode and a cathode are separated is used, and electrolysis is carried out while emulsifying the organic halide-containing liquid using ultrasonic wave generating means, at the same time using the cavitation effect of ultrasonic waves, which forms at normal temperature and normal pressure a vacuum state with a pressure of 1000 atm and a temperature of 5000°C, and stirring the liquid by stirring means. In this case, a method in which the organic halide or the liquid containing the organic halide is put into the cathode side, and electrolysis is carried out while applying ultrasonic waves to the cathode side using ultrasonic wave generating means and stirring on the cathode side by stirring means is more efficient. The application of ultrasonic waves and the stirring are not necessarily carried out all the time, and they may be carried out intermittently according to the situation.

[0014] The organic halide that is targeted in the present invention means an aliphatic, alicyclic, aromatic, or polycyclic compound having fluorine, chlorine, bromine, or iodine in the molecule. For example, PCBs and dioxins, which are considered to cause problems that affect the environment, are representative

examples.

[0015] Electrolysis conditions depend on the state of the target organic halide or a liquid containing the organic halide, but they are usually adjusted to give a voltage range of 5 to 500 V and a current range of 5 to 100 A. However, when the electrolytic cell is large, electrolysis conditions in which the voltage and current are higher are employed accordingly. For electrolysis, use of a direct current power supply is appropriate, but an alternating current power supply of, for example, 50 to 60 Hz, or a high frequency power supply of, for example, 1 KHz may be used. When carrying out electrolysis, in order to increase the electric conductivity, it is preferable to carry out electrolysis by adding to the organic halide, the liquid containing the organic halide, or a liquid to be treated containing the above, a salt such as a hydroxide or a chloride of an alkali metal or an alkaline earth metal. By so doing, a voltage of, for example, 200 V necessary for water is reduced to 1/10, that is, to about 20 V, and the temperature increase of the liquid that is treated can be suppressed, which is desirable. The amount thereof added is 1 to 50 g per L of the liquid to be treated, and preferably 3 to 10 g. It is preferable to carry out electrolysis under conditions in which a hydroxide or chloride salt is added to the anode side and a smaller amount thereof or none is added to the cathode side.

[0016] The present invention carries out, as a reduction reaction at the electrolysis cathode, a substitution reaction between a halogen of the organic halide and hydrogen generated by electrolysis of water, etc. However, if the cathode and the anode are not separated by

a diaphragm, a reversible reaction proceeds, and there is a possibility that a PCB might be regenerated at the anode as a result of an oxidation reaction. It is therefore desirable to prevent the reversible reaction by preventing the product at the cathode from physically contacting the anode. As a method therefor, a diaphragm electrolytic cell in which an anode chamber and a cathode chamber are separated is used. As the diaphragm, there are an ion-exchange membrane, an organic or inorganic microporous membrane, etc., and an appropriate one may easily be selected therefrom while taking into consideration corrosion resistance, mechanical strength, pore size and distribution, electrical resistance, etc. The shape is not particularly limited and may be any shape as long as it prevents substances that have been generated or are present at the anode and the cathode and have dissolved in an electrolyte from being mixed by diffusion convection. In practice, a diaphragm electrolytic cell having a configuration in which an anode in the electrolytic cell is covered with a cylindrical anion-exchange membrane is convenient.

[0017] In the present invention, an organic halide or a liquid containing the organic halide is used in electrolysis as it is or in the form of an organic solvent solution or a mixed liquid (liquid to be treated) with an aqueous emulsion or water. For example, when a PCB is an insulating oil of a capacitor, etc., it may be made into a mixed liquid by adding an appropriate amount of water, made into an emulsion using an appropriate surfactant, or emulsified by, for example, a treatment with ultrasonic waves or microwaves. The same applies in the case of a dioxin, and in the case of, for example, soil contaminated by a dioxin, an appropriate amount of water is added together with a surfactant as necessary, the soil is suspended by mixing while stirring, and it is then subjected to electrolysis as it is or, by removing solids, as a liquid for treatment. When carrying out a reaction, it is of course possible to stir the liquid to be treated by electrical or mechanical stirring means.

[0018] In the present invention, when carrying out a dehalogenating reaction through electrolysis, ultrasonic waves are applied to the reaction system using ultrasonic wave generating means such as, for example, an ultrasonic wave generating apparatus. In accordance with the use of such an operation in combination with electrolysis, not only is the cavitation effect due to ultrasonic waves exhibited in the reaction liquid, but also, due to the emulsion effect of ultrasonic waves, micro clusters are formed from the organic halide or from the organic halide and another liquid, thus promoting the reaction more efficiently. As the ultrasonic wave generating means, for example, a known or commercially available ultrasonic wave generation apparatus may be employed. The power of the ultrasonic waves is on the order of 1 to 100 W per L of the liquid to be treated, and preferably 10 to 40 W. In addition to the so-called ultrasonic waves, the ultrasonic wave generating means referred to in the present invention includes means that imparts vibration to an object

by frequency such as microwaves.

[0019] It is generally convenient to employ room temperature for the reaction temperature, but it may be adjusted in a range from the lowest temperature at which the organic halide or the liquid containing the organic halide does not freeze to the reflux temperature. Furthermore, for example, when electrolysis is carried out at high voltage, the temperature of the liquid to be treated increases, but in such a case the electrolytic cell may be cooled and the liquid to be treated that has been evaporated may be passed through a cooling apparatus and returned to the electrolytic cell. The reaction time depends on the amount to be treated, but it is usually adjusted so as to be in the range of 10 to 100 minutes. It is unnecessary to pay special attention to the reaction atmosphere, and the reaction system may be an open system at normal pressure, but in order to eliminate the influence of air as much as possible, the reaction may be carried out under an inert atmosphere such as nitrogen.

[0020] The electrolysis of the present invention is carried out using an electrolytic cell for carrying out dehalogenation by subjecting an organic halide or a liquid containing the organic halide to electrolysis, the electrolytic cell comprising an anode and a cathode separated by the above-mentioned diaphragm, ultrasonic wave generating means, and stirring means. Examples of the anode include platinum, titanium, and carbon. Examples of the cathode include metals such as palladium and titanium, and alloys thereof. The ultrasonic wave generating means may be any type as long as the liquid to be treated is sufficiently emulsified by the emulsion effect and dehalogenation is promoted by the cavitation effect, but it is suitable to use a type in which an ultrasonic wave oscillator rod connected to an ultrasonic wave generator is inserted into the liquid to be treated. The stirring means may employ a known or commercially available stirrer. As a treatment vessel, it is unnecessary to use a special vessel, and in practice electrolysis may be carried out using a general container and apparatus made of, for example, stainless steel, glass, or polyvinyl chloride.

[0021] Another embodiment of the present invention is a method in which, when carrying out a dehalogenation reaction by electrolyzing an organic halide or a liquid containing the organic halide, a hydrogen-absorbing metal or alloy is used as a cathode. It is preferable to use a hydrogen-absorbing metal such as palladium or a hydrogen-absorbing alloy such as a titanium-iron alloy as the cathode since hydrogen generated by electrolysis is absorbed by the cathode, subsequently the hydrogen discharged therefrom efficiently replaces a halogen, thus contributing to the dehalogenation reaction, and a reduction reaction is thereby carried out efficiently.

[0022] In this case also, it is preferable to employ a method in which a diaphragm electrolytic cell is used for electrolysis, and a more preferred result can be obtained by employing a method in which electrolysis is carried out while applying ultrasonic waves to the liquid to be treated and stirring using ultrasonic wave generating

means and stirring means.

[0023] Yet another embodiment of the present invention is an electrolytic cell for carrying out dehalogenation by electrolyzing an organic halide or a liquid containing the organic halide, the electrolytic cell comprising an anode and a cathode separated by a diaphragm, ultrasonic wave generating means, and stirring means. The ultrasonic wave generating means and the stirring means may be provided on both the cathode side (cathode chamber) and the anode side (anode chamber), and providing them at least on the cathode side (cathode chamber) is efficient.

[0024] An electrolytic cell used for carrying out the electrolysis of the present invention is explained by reference to a drawing. FIG. 1 is a schematic diagram of an electrolytic cell as one example thereof. In FIG. 1, an anode 1 (platinum electrode) and a cathode 2 (palladium electrode) are separated by a diaphragm 3 (anionic membrane). A liquid to be treated 6 is also divided into an anode chamber 4 and a cathode chamber 5 by the diaphragm 3, and from the viewpoint of efficiency a liquid to be treated comprising an organic halide or a liquid containing the organic halide is placed in the cathode chamber 5, and an aqueous solution of a salt such as a hydroxide or a chloride of an alkali metal or an alkaline earth metal is placed in the anode chamber 4. An ultrasonic wave oscillator rod 7 for applying ultrasonic waves to the liquid to be treated 6 is inserted into the cathode chamber 5, and stirring means 8 is placed in the cathode chamber 5. The ultrasonic wave oscillator rod 7 and the stirring means 8 may be inserted into/placed in both chambers. By carrying out electrolysis using the above-mentioned electrolytic cell while applying ultrasonic waves and passing current between the anode and the cathode, a dehalogenation reaction of the organic halide proceeds.

[0025] The present invention is explained in detail below by reference to Examples. In the Examples, the concentrations of PCB and dioxin were measured by a GC/MAS method, which is normally employed as an analytical method therefor.

Example

Example 1

[0026] A liquid to be treated was prepared by mixing 1 L (liter) of a capacitor liquid containing a PCB (PCB concentration, 100 ppm), 10 g of sodium hydroxide, and 20 L of water. The electrolytic cell (treatment vessel being made of stainless steel) of FIG. 1 was used, this liquid to be treated was placed in the cathode chamber, 20 L of an aqueous solution containing 100 g of sodium hydroxide was placed in the anode chamber, a three-phase direct current power source was used as a power source, and electrolysis was carried out at normal temperature and normal pressure for 30 minutes while applying ultrasonic waves. The average voltage of the electrolysis was 20 V and the average current was 50 A. An electric wave

rod, which corresponded to the ultrasonic wave generating means, was a metal rod having a diameter of 45 mm and a length of 35 cm, and ultrasonic waves of 20 KHz were applied thereto. The power of the ultrasonic waves was on average 255 W. The applied voltage during electrolysis was substantially constant for 30 minutes, but the current and the power of the ultrasonic waves gradually increased. The liquid to be treated in the cathode chamber was stirred at 1400 revolution/min using a commercial mixer. The PCB concentration in the treated liquid after the electrolysis was 0.5 ppm or less, which was considerably less than the 100 ppm prior to the electrolysis.

Example 2

[0027] 20 L of an aqueous liquid to be treated containing 500 pg/L of a dioxin was electrolyzed in the same manner and using the same electrolytic cell as in Example 1. The dioxin concentration in the treated liquid after the electrolysis was 0.5 pg or less.

[0028] In accordance with the method of the present invention, transformer oil containing PCBs, etc. or another liquid containing PCBs, etc., which have been causing problems in recent years, or incinerated ash or soil containing dioxins can be disposed of by a relatively simple method and apparatus, that is, very economically. Moreover, since the operation is carried out at normal temperature and normal pressure, they can be made harmless by a safe and clean treatment employing water and electricity, and the application value is very high from industrial and environmental viewpoints.

Claims

1. A method for dehalogenating an organic halide, wherein when carrying out dehalogenation by electrolyzing the organic halide or a liquid containing the organic halide, a diaphragm electrolytic cell in which an anode and a cathode are separated is used, and electrolysis is carried out while applying ultrasonic waves using ultrasonic wave generating means and stirring using stirring means.
2. The method for dehalogenating an organic halide according to Claim 1, wherein the organic halide is a PCB or a dioxin.
3. The method for dehalogenating an organic halide according to either Claim 1 or 2, wherein electrolysis is carried out by adding to the organic halide or the liquid containing the organic halide a salt of an alkali metal or an alkaline earth metal.
4. A method for dehalogenating an organic halide, wherein when carrying out dehalogenation by electrolyzing the organic halide or a liquid containing the

organic halide, electrolysis is carried out using a hydrogen-absorbing metal or alloy as a cathode.

5. The method for dehalogenating an organic halide according to Claim 4, wherein a diaphragm electrolytic cell in which an anode and the cathode are separated is used.
6. The method for dehalogenating an organic halide according to either Claim 4 or 5, wherein electrolysis is carried out on the organic halide or the liquid containing the organic halide while applying ultrasonic waves using ultrasonic wave generating means and stirring by stirring means.
7. The method for dehalogenating an organic halide according to either Claim 5 or 6, wherein the organic halide or the liquid containing the organic halide is put into the cathode side, and electrolysis is carried out while applying ultrasonic waves to the cathode side using ultrasonic wave generating means and stirring on the cathode side by stirring means.
8. An electrolytic cell for carrying out dehalogenation by electrolyzing an organic halide or a liquid containing the organic halide, wherein the electrolytic cell comprises an anode and a cathode separated by a diaphragm, ultrasonic wave generating means, and stirring means.
9. The electrolytic cell according to Claim 8, wherein the cathode is formed from a hydrogen-absorbing metal or alloy.
10. The electrolytic cell according to either Claim 8 or 9, wherein the ultrasonic wave generating means and the stirring means are provided on the cathode side.

Amended claims under Art. 19.1 PCT

1. (Amended) A method for dehalogenating an organic halide, wherein when carrying out dehalogenation by electrolyzing the organic halide or a liquid containing the organic halide, (1) a diaphragm electrolytic cell in which an anode and a cathode formed from a hydrogen-absorbing metal or alloy, the anode and the cathode being separated by the diaphragm, is used, (2) the organic halide or the liquid containing the organic halide is put into the electrolytic cell on the cathode side, and (3) electrolysis is carried out while applying ultrasonic waves on the cathode side using ultrasonic wave generating means and stirring on the cathode side by stirring means.
2. The method for dehalogenating an organic halide according to Claim 1, wherein the organic halide is a PCB or a dioxin.

3. (Amended) The method for dehalogenating an organic halide according to either Claim 1 or 2, wherein an aqueous solution of a salt such as a hydroxide or a chloride of an alkali metal or an alkaline earth metal is put into the electrolytic cell on the anode side and electrolysis is carried out. 5
4. (Cancelled)
5. (Cancelled) 10
6. (Cancelled)
7. (Cancelled) 15
8. (Amended) A diaphragm electrolytic cell for carrying out dehalogenation by electrolyzing an organic halide or a liquid containing the organic halide, in which an anode and a cathode are separated, wherein the electrolytic cell on the cathode side comprises ultrasonic wave generating means and stirring means, and the cathode is formed from a hydrogen-absorbing metal or alloy. 20
9. (Cancelled) 25
10. (Cancelled)
11. (Newly added) The diaphragm electrolytic cell according to Claim 8, wherein the electrolytic cell comprises a cooling means for the electrolytic cell and a refluxing means for a liquid to be treated that has been evaporated. 30

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Statement under Art. 19.1 PCT

The amended Claim 1 is substantially a combination of the original Claims 1, 4, 5, 6 and 7.

The Claim 2 is retained unchanged. 40

The amended Claim 3 is based on the paragraphs 0017 and 0026 of the description.

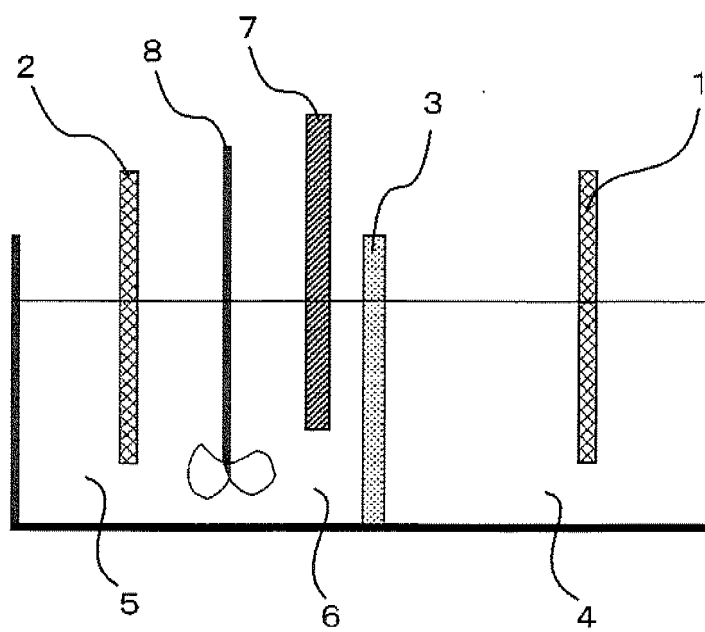
The amended Claim 8 is substantially a combination of the original Claims 8, 9 and 10.

The newly added Claim 11 depends on the Claim 8, and is based on the paragraph 0021 of the description. 45

A feature of "applying ultrasonic waves to the cathode side using ultrasonic wave generating means and stirring on the cathode side by stirring means" in the amended Claim 1 and a feature of "the electrolytic cell on the cathode side comprises ultrasonic wave generating means and stirring means" in the amended Claim 8 are not described in any of five cited documents listed in the international search report. We think that a significance of the requirements is described in the paragraph 0024 of the description, is supported by a drawing and the description of the paragraph 0025 of the description, and can not be expected in view of the descriptions of 50 55

the five cited documents, so that it can be a basis of an inventive step of the invention sufficiently. Further, the feature of the newly added Claim 11 is not described in any of the five cited documents.

FIG. 1



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INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2005/004922

A. CLASSIFICATION OF SUBJECT MATTER Int. Cl. ⁷ A62D3/00 According to International Patent Classification (IPC) or to both national classification and IPC		
B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) Int. Cl. ⁷ A62D3/00 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Jitsuyo Shinan Koho 1922-1996 Jitsuyo Shinan Toroku Koho 1996-2005 Kokai Jitsuyo Shinan Koho 1971-2005 Toroku Jitsuyo Shinan Koho 1994-2005 Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	JP 2002-345991 A (Kenji SUZUKI, Ait Co., Ltd.), 03 December, 2002 (03.12.02), Pages 3 to 4, Claims; pages 4 to 5, Par. No. [0014]; page 5, Par. Nos. [0021], [0028], [0028] & EP 1391223 A1 & WO 2002/094382 A1	1-10
Y	JP 2000-79377 A (PBM Kabushiki Kaisha), 21 March, 2000 (21.03.00), Claims; Par. No. [0014] (Family: none)	1-3, 5-10
Y	JP 2000-80489 A (PBM Kabushiki Kaisha), 21 March, 2000 (21.03.00), Claims; Par. No. [0007] (Family: none)	1-3, 5-10
☒ Further documents are listed in the continuation of Box C. ☐ See patent family annex.		
* Special categories of cited documents: "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier application or patent 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 when the document is taken alone "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		
Date of the actual completion of the international search 02 June, 2005 (02.06.05)		Date of mailing of the international search report 21 June, 2005 (21.06.05)
Name and mailing address of the ISA/ Japanese Patent Office		Authorized officer
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Form PCT/ISA/210 (second sheet) (January 2004)

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2005/004922

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	JP 2003-89893 A (Sharp Corp.), 28 March, 2003 (28.03.03), Claims; Par. No. [0023] (Family: none)	4-7, 9, 10
Y	JP 11-90449 A (Permelec Electrode Ltd.), 06 April, 1999 (06.04.99), Claims (Family: none)	4-7, 9, 10

Form PCT/ISA/210 (continuation of second sheet) (January 2004)

REFERENCES CITED IN THE DESCRIPTION

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- JP 2001269673 A [0006]
- JP 8066494 A [0006]
- JP 2002265391 A [0006]
- JP 2002345991 A [0006]