

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AT	Austria	ES	Spain	MG	Madagascar
AU	Australia	FI	Finland	ML	Mali
BB	Barbados	FR	France	MN	Mongolia
BE	Belgium	GA	Gabon	MR	Mauritania
BF	Burkina Faso	GB	United Kingdom	MW	Malawi
BG	Bulgaria	GN	Guinea	NL	Netherlands
BJ	Benin	GR	Greece	NO	Norway
BR	Brazil	HU	Hungary	PL	Poland
CA	Canada	IT	Italy	RO	Romania
CF	Central African Republic	JP	Japan	SD	Sudan
CG	Congo	KP	Democratic People's Republic of Korea	SE	Sweden
CH	Switzerland	KR	Republic of Korea	SN	Senegal
CI	Côte d'Ivoire	LI	Liechtenstein	SU	Soviet Union
CM	Cameroon	LK	Sri Lanka	TD	Chad
CS	Czechoslovakia	LU	Luxembourg	TG	Togo
DE	Germany	MC	Monaco	US	United States of America
DK	Denmark				

1 the exudation of species of pine, and containing abietic
acid and its anhydride. Typically, a small amount of
another acid is added to rosin flux to activate it and
such compositions are referred to as "acid rosin flux" or
5 "activated rosin flux." After the solder operation has
been completed, excess flux as well as flux residues
remain on the soldered surface, and these residues are
resinous, waxy and conductive. These flux residues and
excess flux must be removed prior to subsequent processing
10 steps in order to prevent reaction thereof with the bonded
part, leading to corrosion and resultant electrical
insulation losses.

Defluxing solvents which are widely used at present
are halogenated hydrocarbons such as 1,1,1-trichloroethane
15 and Freon (a tradename of E. I. DuPont for polyhalogenated
hydrocarbons including chlorine and fluorine). While
these organic materials are effective defluxing solvents,
they have the serious disadvantage that they have a
negative impact on the environment due to air pollution
20 and ozone depletion. In fact, recent environmental
legislation mandates that these materials be banned or
their production severely restricted in the next few
years. When these materials are used, even in small
quantities, expensive management systems for transport,
25 storage, use, and disposal and environmental protection
equipment must be used to prevent air and water
discharges. In addition, waste solvents require energy
intensive regeneration operations for these materials.

While numerous aqueous cleaners are available which
30 are viable degreasing solvents, none of these has been
found to be effective as an electronic component defluxing
solvent. In addition, the resulting organic-laden aqueous
solvents require further processing before disposal.

Thus, an urgent need exists in the electronics
35 industry for a solvent which effectively removes organic

1 flux residues while at the same time avoiding any negative
environmental impact. Such a solvent would also be
desirable for removing other organic materials from other
substrates.

5

SUMMARY OF THE INVENTION

The general purpose of the present invention is to
provide a new and improved composition and method for
10 removing organic contaminants from a chosen solid
substrate while avoiding negative environmental impact.
This composition and method possess the advantages of the
above prior art processes while overcoming their
significant disadvantages.

15 The above general purpose of this invention is
accomplished by first providing a composition comprising:
(a) hydrogen peroxide in the amount of about 3 to 5 percent
by weight of said composition; (b) an alkaline compound in
sufficient amount to provide a pH of at least 10.5 in said
20 composition; (c) a chosen wetting agent in the amount of
about 0.1 to 0.3 percent by weight of said composition,
said wetting agent being unreactive with said hydrogen
peroxide and said alkaline compound; and (d) purified
water as the balance of said composition.

25 Optionally, the composition may further comprise a
chosen metal protective agent in the amount of about 0.5
to 2.0 percent by weight of said composition, said metal
protective agent being unreactive with said hydrogen
peroxide and said alkaline compound.

30 The solid substrate having organic contaminants
thereon is exposed to the above-noted composition whereby
the organic contaminants are removed from the substrate
and are converted into non-toxic and non-hazardous
products. Thus, negative environmental impact is avoided
35 by the present process. In an alternative embodiment of

1 the present invention, the organic contaminant removal is
further enhanced by exposing the composition and the
organic contaminants on the substrate to ultraviolet
radiation.

5

DESCRIPTION OF THE PREFERRED EMBODIMENTS

The composition of the present invention is effective
for removing organic contaminants from a solid substrate
10 while at the same time avoiding undesirable impact on the
environment. The composition of the present invention is
especially useful for removing flux residues which
contaminate surfaces after a soldering operation. These
flux residues typically comprise resinous, waxy
15 contaminants which are the breakdown products from the
soldering operation. The substrates which are soldered
comprise, for example, printed wiring boards, integrated
circuits, electronic components, electronic devices,
electronic connectors, or electronic cables. In
20 accordance with the present invention, the by-products,
such as carbon dioxide, nitrogen, and water, which are
formed are non-hazardous and can be removed without having
a negative environmental impact.

The composition in accordance with the present
25 invention comprises: (a) hydrogen peroxide (H_2O_2) in
the amount of about 3 to 5 percent by weight; (b) an
alkaline compound in a sufficient amount to provide a pH
of 10.5 or higher in the composition; (c) a chosen wetting
agent which is unreactive with hydrogen peroxide and the
30 alkaline compound and which is present in the amount of
about 0.1 to 0.3 percent by weight of the composition; and
(d) purified water as the balance of the composition.
Optionally, if the substrate comprises metal, a
metal-protective agent is added to protect the metal
35 surface from attack by the peroxide and alkali. The metal

1 protective agent is unreactive with the hydrogen peroxide
and the alkaline compound and is present in the amount of
about 0.5 to 2.0 percent by weight of the composition.

5 The hydrogen peroxide acts as a solvent, emulsifier,
and oxidant. While not limiting the present invention to
a particular theory of operation, it is believed that the
hydrogen peroxide has the following effect. After being
immersed in the present composition, a flux-contaminated
10 substrate invokes selective adsorption of hydrogen
peroxide into the flux film and rapid decomposition of the
inorganic peroxide. The selective adsorption of hydrogen
peroxide by the rosin-based contaminant films is due to
the ether-like solubility chemistry of hydrogen peroxide.
Following adsorption, hydrogen peroxide rapidly decomposes
15 into water and oxygen gas. The oxygen emulsifies the
resinous contaminant, increasing surface area and
solvency. This adsorption and emulsification process is
observed as spontaneous foaming on the contaminated
surface. The scrubbing foam enhances the cleaning
20 activity of the solvent, dramatically improving
penetration of solvent into low-tolerance spacings. After
the organic flux contaminants have been removed from the
substrate, they are oxidized by the action of nascent or
atomic oxygen which is formed by the spontaneous
25 decomposition of hydrogen peroxide, and are decomposed
into carbon dioxide, nitrogen, and water. This effect is
evidenced by a change in color in the composition from
amber when it contains dissolved flux, to clear. The
contaminants may be dissolved or suspended material.

30 The effectiveness of the composition of the present
invention is believed to be due to a synergistic
relationship between pH, peroxide chemistry and the
wetting agent. It was found that peroxide-based
formulations with pH's of 2 (acidic) and 7 (neutral) did
35 not effectively remove contaminants. In these cases, the

1 flux contaminants tended to gel and discolor on the
substrate surfaces. Alkaline formulations having a pH of
greater than 10.5 were considerably more effective at
solubilizing the flux residues than acid or neutral
5 solutions. (This is probably due to ionization of flux
acids by basic solutions, forming primitive soaps.) Thus,
the alkaline compound is added to the present composition
to provide a pH of at least 10.5, preferably within the
range of 10.5 to 11.5. The alkaline compound may be, for
10 example, sodium hydroxide or potassium hydroxide, with
sodium hydroxide being most preferred. If sodium
hydroxide is used, it is present in the amount of about
0.2 to 0.5 percent by weight of the composition.

The wetting agent must be chosen to be compatible with
15 the other components in the present composition.
Conventional wetting agents such as sodium alkylaryl
sulfonate and other organic synthetic detergents decompose
rapidly in the presence of strong alkaline oxidizing/
bleaching solutions such as the composition of the present
20 invention. This results in rapid decomposition of the
hydrogen peroxide solvent and excessive foaming in the
solution. A preferred wetting agent for use in the
present composition comprises sodium 2-ethylhexyl sulfate
(obtained from Niacet Co., Niagara Falls, New York). It
25 has good solubility, stability, and penetrating action in
near-boiling alkaline and acid solutions, and is one of
the few anionic surfactants stable in concentrated
bleaching solutions. Other suitable wetting agents for
practising the present invention may comprise sodium
30 metasilicate or short-chain branched surfactants. The
wetting agent is used in the present invention in the
amount of about 0.1 to 0.3 percent by weight of the
composition.

If the substrate comprises a metal, such as in a
35 printed wiring board, the metal surfaces must be protected

1 from attack by the peroxide and alkali in the present
composition. The particular metal protective agent used
depends on the specific metal being protected. For
example, calcium and phosphorous compounds are used as
5 protective agents for copper. The selection of the
particular protective agent for a particular metal is
known in the art and will not be detailed here. Suitable
metal protective agents for practising the present
invention include sodium carbonate or sodium
10 metasilicate. The metal protective agent is present in
the amount of about 0.5 to 2.0 percent by weight of the
composition.

The optimum operating temperature of the present
composition to provide effective contaminant removal
15 within a short period of time is within the range of 51.7
to 71.1°C (125 to 160°F). Within this range, flux
contaminants may be removed in 1 to 3 minutes. A
preferred temperature for use of the present composition
is about 60°C (140°F). The increased temperature promotes
20 the reaction, improves penetration of the composition by
softening gummy contaminants, and keeps particulates in
suspension so that they do not deposit on the cleaned
substrates. In addition, the elevated temperature
optimizes the effectiveness of the ultraviolet radiation
25 for cleaving bonds in organic materials, as described
below.

The contaminants which can be removed in accordance
with the present invention comprise organic materials
which include, but are not limited to the residues left by
30 commonly used flux materials. These flux residues
comprise oils, resins, and other organic materials. Since
the present composition and method are effective for
cleaving carbon-to-carbon bonds, they may be used for the
removal of other organic compounds as well. Such
35 materials include, but are not limited to, oil, grease,

1 lubricants, photoresist, adhesive residues, plasticizers,
or dyes.

The solid substrate from which contaminants can be removed in accordance with the present invention may
5 comprise any material which is not adversely affected by the peroxide or alkaline components of the present composition. Such materials include, but are not limited to, polyimide/ polyamide laminates and epoxy/glass laminates which are used in printed wiring boards, silicon
10 which is used in electronic devices, and anodized aluminum or polyimide which are used in cables and connectors. The solid substrate may have a simple or complex configuration and may include interstitial spaces which are difficult to clean by known methods. The substrate may be in the form
15 of a continuous layer or in the form of discrete particles.

In accordance with an alternative embodiment of the present invention, ultraviolet radiation is used to enhance the cleaning process. When hydrogen peroxide is exposed to ultraviolet radiation having a wavelength
20 within the range of 184 to 300 nanometers (nm), preferably about 253 nm, such as from a xenon flash lamp, the hydrogen peroxide is dissociated to form a hydroxyl radical (OH^-) which is very reactive. This hydroxyl radical then cleaves the carbon-to-carbon bonds in the
25 organic contaminant material, forming carbon dioxide and water. In addition, the ultraviolet radiation itself also cleaves the carbon-to-carbon bonds in the organic contaminant materials, adding even further effectiveness to the cleaning process. The source of ultraviolet
30 radiation, such as a xenon flash lamp or a mercury vapor lamp, is located external to the chamber containing the substrate to be cleaned and the present composition, and the radiation is directed into the cleaning chamber. For this purpose, a quartz window is provided on one surface
35 of the cleaning chamber to permit transmission of the

1 ultraviolet radiation. Optionally, a focusing barrel may
 be provided between the quartz window and the radiation
 source to improve the efficiency of the ultraviolet
 radiation transmission. Such methods for introducing
 5 radiation into reaction chambers are well known.

The composition of the present invention may be used
 in essentially the same manner as known defluxing agents
 are used, namely in a spray or soak operation. If used in
 a soak operation, it is desirable to keep the present
 10 composition agitated, by air or mechanical or ultrasonic
 means. As an added feature in the present method, after
 the substrate has been treated with the present
 composition, residual alkaline materials (alkali or alkali
 salts) remaining on the substrate are neutralized. A
 15 preferred neutralization agent comprises hot carbonated
 water, which is applied at 51.7 to 71.1°C (125 to 160°F).
 The carbonated water may be formed by bubbling carbon
 dioxide into deionized water. The use of the carbonated
 water rinse keeps the rinse water below a pH of 9.5, which
 20 is the regulated limit for disposal in a sewer. After the
 neutralization step, the substrate is rinsed in hot water
 at 51.7 to 71.1°C (125 to 160°F), preferably with air
 agitation, and then dried, preferably with hot air. In an
 exemplary batch processing technique, in which
 25 contaminated substrates are loaded into a chamber and
 treated, in turn, with the present composition,
 neutralizing agent, water, and hot air, the following
 approximate processing times may typically be used:

30	<u>Step</u>	<u>Time</u>
a.	Exposure of contaminated substrate to present composition	3 minutes
35	b. Neutralization	3 minutes
	c. Hot water rinse	4 minutes
	d. Hot air drying	5 minutes

1 Thus, the present process can be completed within 15
minutes.

Alternatively, the present process may be practised as
an "in-line" process. The contaminated substrates are
5 loaded into a rack and the rack is lowered in sequence
into a series of chambers containing, respectively, the
present composition, the neutralizing agent, and deionized
water sprayers. As yet another alternative, this in-line
processing may be accomplished by a conveyORIZED system.

10 After the substrate has been treated with the present
composition, the substrate is removed from the chamber
containing the composition. The starting composition is
then re-established by the addition of hydrogen peroxide
and, if necessary, the other components of the
15 composition. Thus, the bulk cleaning solvent remains in
place and does not require disposal. Rather, the
composition is regenerated in situ by the addition of
component materials as required.

As an added feature of the present invention, the
20 present composition removes metallic oxides from the
substrate and thus serves also as a solder brightener.
This eliminates the need for a secondary process, such as
treatment with fluoroboric acid.

25 Examples of practise of the present invention are as
follows.

EXAMPLE 1

30 This example illustrates the use of the composition
and method of the present invention to remove solder flux
from printed wiring boards.

It should be noted that while the present composition
is non-toxic, general safety precautions, such as the use
of goggles and protective clothing, are necessary.

1 The test samples comprised polyimide/polyamide
lamine printed wiring boards containing copper
metallization and having been exposed to Alpha 611, a
solder flux obtained from Alpha Metals Company of
5 Alpharetta, Georgia, and comprising a mildly activated
rosin flux. The composition of the present invention
comprised: approximately 3.0 percent by weight hydrogen
peroxide; approximately 0.2 percent by weight sodium
hydroxide; approximately 0.1 percent by weight sodium
10 2-ethylhexyl sulfate as the wetting agent; approximately
0.5 percent by weight sodium metasilicate as the metal
protective agent; and the balance being purified water.
The temperature of the composition was maintained at 60°C
(140°F). The samples were treated in a batch process as
15 previously described herein using the previously described
processing steps and times. The present composition was
nearly transparent prior to introduction of flux-
contaminated materials, with only trace amounts of gas
liberation visible. When the contaminated material was
20 introduced, vigorous gas formation on the flux residue was
observed. This action continued as the flux was
emulsified and dissolved into solution. Continued gas
formation was visible as the dissolved flux residues
continued to be oxidized. Completion of the decomposition
25 was indicated by slow gas evolution and change in fluid
color. Surface foaming was minimal. Complete removal of
the flux residues was accomplished in an average of less
than two minutes. The soldered joints were clean and free
of oxides, i.e., bright in appearance.

30 By contrast, it was found that alkaline solutions
alone did not effectively separate the bulk of the flux
residues from the substrate. In addition, alkaline
solutions with wetting agents, but without hydrogen
peroxide, required in excess of 10 minutes to separate

1 flux residues from the substrate. Even after separation,
the flux residues did not disperse well.

EXAMPLE 2

5

This example illustrates the use of the composition and method of the present invention to remove solder flux from cable connectors.

10 A soft anodized aluminum connector was treated with the composition of the present invention as described in Example 1 except using a temperature of 160°F (71°C) and 60-second rinse times for both the neutralization and final rinse steps. There was no visually detectable degradation of the anodized aluminum by exposure to the
15 present composition.

In another experiment, a cable made of Kapton (a trademark of E.I. du Pont de Nemours for a polyimide) was contaminated with Kester 197, a mildly activated rosin based flux obtained from Litton-Kester Company of Chicago, Illinois. The Kapton cable was treated in accordance with
20 the present invention as described above. Complete removal of the flux was accomplished as determined by visual inspection under 25X magnification.

25

EXAMPLE 3

This example illustrates the use of the embodiment of the method of the present invention in which ultraviolet radiation is used to enhance the contaminant removal.

30 The composition and method described in Example 1 are used except that the composition and the substrate containing contaminants are exposed to radiation from a xenon flash lamp as previously described herein. The cleaning process proceeds generally as described in

1 Example 1, except that complete flux removal is
accomplished more rapidly.

As previously discussed, the organic contaminants are
oxidized by the present composition to form carbon
5 dioxide, nitrogen, and water. The carbon dioxide and
nitrogen are non-hazardous and may be exhausted into the
atmosphere. The water by-product contains no noxious
material and may be disposed of in a sewer without further
treatment or may be re-used. The hydrogen peroxide in the
10 present composition is decomposed during the cleaning
operation into atomic oxygen or hydroxyl radicals and
water and the former react with the contaminants to form
the above-noted products. Insoluble precipitates, such as
cellulose gums, may also be formed as by-products of the
15 bond cleaving of the present composition, and may be
readily removed by filtration. Thus, no toxic or
hazardous materials are formed as a result of the present
method. Consequently, the present process obviates the
need for the expensive solvent emission control and waste
20 management procedures required when using prior art
halogenated solvents. In addition, environmental
pollution and exposure of workers to hazardous materials
are also avoided by the present composition and method.

While the previous discussion has focused on the use
25 of the present composition and method to remove flux
residues from solid substrates, it is not intended to
limit the present invention to this particular
contaminant. Rather, it is intended to include within the
scope of the present invention the removal of any organic
30 material from a given solid substrate.

Having thus described exemplary embodiments of the
present invention, it should be noted by those skilled in
the art that the within disclosures are exemplary only and
that various other alternatives, adaptations and

1 modifications may be made within the scope of the present invention. Accordingly, the present invention is not limited to the specific embodiments as illustrated herein, but is only limited by the following claims.

CLAIMSWhat is Claimed is:

- 1 1. A composition for removing organic contaminants
from a chosen solid substrate by decomposing said
contaminants into non-hazardous by-products, said
composition comprising:
- 5 (a) hydrogen peroxide in the amount of about 3 to 5
percent by weight of said composition;
- (b) a metal hydroxide in sufficient amount to
provide a pH of at least 10.5 in said composition;
- 10 (c) a wetting agent in the amount of about 0.1 to
0.3 percent by weight of said composition, said wetting
agent being unreactive with said hydrogen peroxide and
said metal hydroxide; and
- (d) purified water as the balance of said
composition.
- 1 2. The composition of Claim 1 wherein said metal
hydroxide is sodium hydroxide or potassium hydroxide.
- 1 3. The composition of Claim 1 further comprising a
metal protective agent in the amount of about 0.5 to 2.0
percent by weight of said composition, said metal
protective agent being unreactive with said hydrogen
5 peroxide and said metal hydroxide.
- 1 4. The composition of Claim 3 wherein:
- (a) said hydrogen peroxide is present in the amount
of about 3.0 percent by weight of said composition;
- 5 (b) said metal hydroxide comprises sodium hydroxide
and is present in the amount of about 0.2 percent by
weight of said composition;

(c) said wetting agent comprises sodium 2-ethylhexyl sulfate and is present in the amount of about 0.1 percent by weight of said composition;

10 (d) said metal protective agent comprises sodium metasilicate and is present in the amount of about 0.5 percent by weight of said composition.

1 5. A method for removing organic contaminants from a chosen solid substrate while simultaneously avoiding negative environmental impact, said method comprising:

5 (a) providing the composition of Claim 1 or Claim 3; and

(b) exposing said solid substrate having said organic contaminants to said composition whereby said organic contaminants are removed from said substrate and converted into non-toxic and non-hazardous products.

1 6. The method of Claim 5 wherein said exposing is performed with said composition at a temperature within the range of 51.7 to 71.1°C (125 to 160°F).

1 7. The method of Claim 5 further comprising after said exposing said substrate to said composition, neutralizing residual said metal hydroxide or products thereof on said substrate, rinsing said substrate with
5 water, and drying said substrate.

1 8. The method of Claim 7 wherein said neutralizing comprises exposing said substrate to hot carbonated water.

1 9. The method of Claim 5 wherein said exposing in step "b" further comprises exposing said composition and said organic contaminants on said substrate to ultraviolet radiation having a wavelength within the range of 184 to
5 300 nanometers.

INTERNATIONAL SEARCH REPORT

International Application No PCT/US 90/06464

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) ⁴ According to International Patent Classification (IPC) or to both National Classification and IPC IPC ⁵ : C 23 G 1/14, C 11 D 7/18		
II. FIELDS SEARCHED Minimum Documentation Searched ⁷		
Classification System	Classification Symbols	
IPC ⁵	C 23 G, C 11 D	
Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included In the Fields Searched ⁸		
III. DOCUMENTS CONSIDERED TO BE RELEVANT⁹		
Category ¹⁰	Citation of Document, ¹¹ with indication, where appropriate, of the relevant passages ¹²	Relevant to Claim No. ¹³
X	EP, A, 0174610 (HENKEL KGaA) 19 March 1986 see claim 4; page 5, lines 18-28; page 9, lines 13-20; page 6, lines 2-10 --	1-5
A	Database World Patent Index, File Server, Accession Number 85-234441, week 38, 1985, Derwent Publication Ltd, (London, GB), & JP, A, 60153982 (TOSHIBA K.K.) 13 August 1985 see abstract --	9
A	EP, A, 0009839 (UNILEVER N.V.) 16 April 1980 --	
A	WO, A, 8809368 (G. SCHAMSCHULA) 1 December 1988 --	
	./.	
<p>¹⁰ Special categories of cited documents:</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"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)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> <p>"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</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"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.</p> <p>"A" document member of the same patent family</p>		
IV. CERTIFICATION		
Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report	
17th April 1991	30 MAY 1991	
International Searching Authority	Signature of Authorized Officer	
EUROPEAN PATENT OFFICE	MISS T. TAZELAAR	

III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)		
Category *	Citation of Document, ** with indication, where appropriate, of the relevant passages	Relevant to Claim No.
A	Patent Abstracts of Japan, vol. 12, no. 223 (C-507)(3070), 24 June 1988, & JP, A, 6320489 (TADANOBU OKUBO) 28 January 1988	
	--	
A	DE, A, 3215966 (LICENTIA PATENT-VERWAL- TUNGS-GmbH) 3 November 1983	

**ANNEX TO THE INTERNATIONAL SEARCH REPORT
ON INTERNATIONAL PATENT APPLICATION NO.**

US 9006464
SA 43302

This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report. The members are as contained in the European Patent Office EDP file on 27/05/91. The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP-A- 0174610	19-03-86	DE-A- 3433593 US-A- 4612142	20-03-86 16-09-86
EP-A- 0009839	16-04-80	AU-A- 5122779 US-A- 4304762	03-04-80 08-12-81
WO-A- 8809368	01-12-88	EP-A- 0314750	10-05-89
DE-A- 3215966	03-11-83	None	