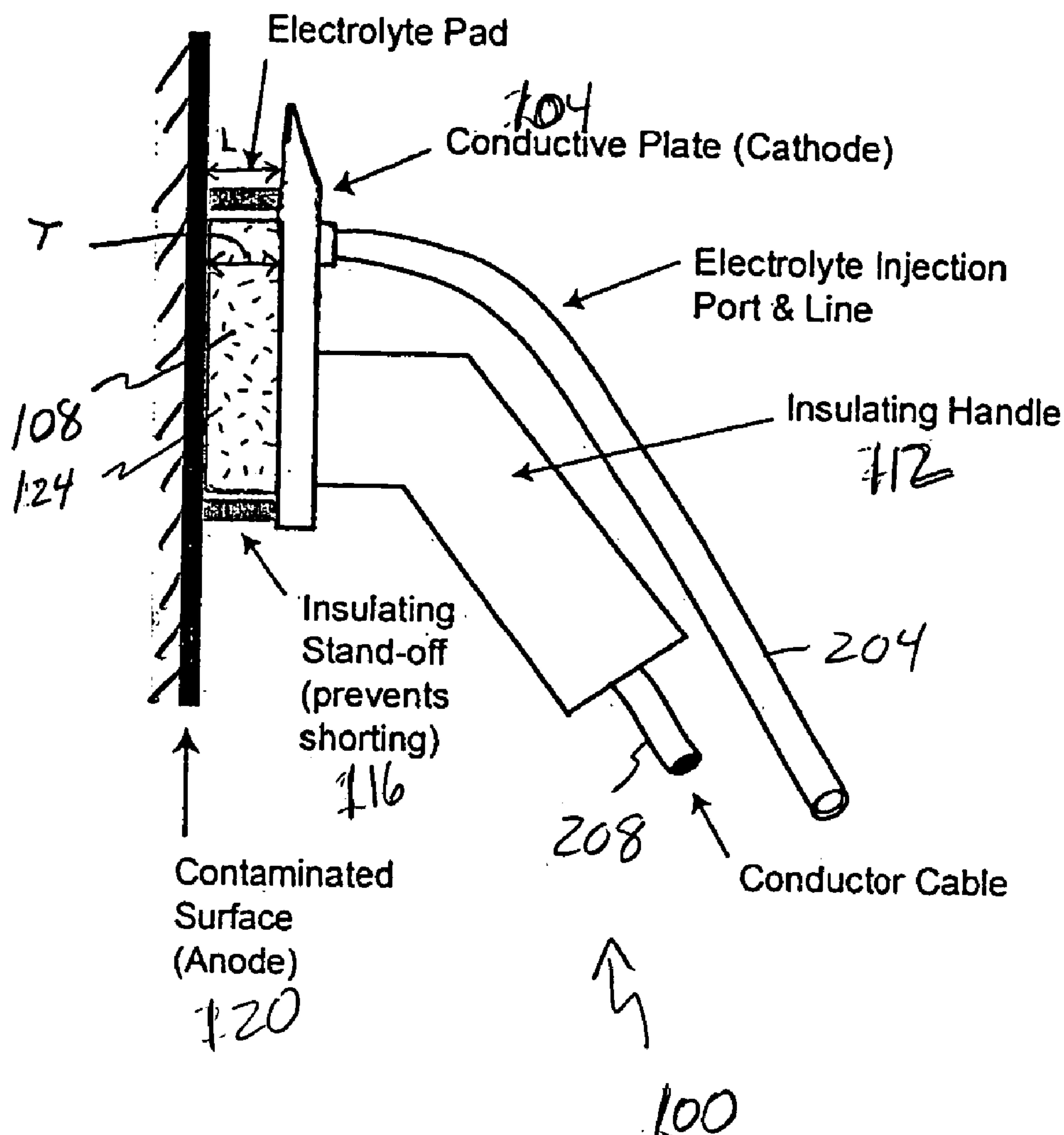


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Veatch et al.(10) **Pub. No.: US 2005/0230267 A1**(43) **Pub. Date: Oct. 20, 2005**(54) **ELECTRO-DECONTAMINATION OF
CONTAMINATED SURFACES****Publication Classification**(76) Inventors: **Bradley D. Veatch**, Westminster, CO
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DENVER, CO 80202(21) Appl. No.: **10/887,576**(22) Filed: **Jul. 9, 2004****Related U.S. Application Data**(60) Provisional application No. 60/486,493, filed on Jul.
10, 2003.(57) **ABSTRACT**

A decontaminating system **200** is provided that includes: (a) a decontaminating apparatus **100** including (i) an electrically conductive scrubbing shoe **104** having an inlet **204** for a gel-like material and (ii) an insulating standoff **116** positioned between the scrubbing shoe **104** and a surface **120** to be decontaminated to maintain a desired distance between the scrubbing shoe **104** and the surface **120**; (b) a reservoir **216** for the gel-like material **124**, the reservoir **216** being in communication with the inlet **204**; and (c) a voltage source **224** in communication with the electrically conductive scrubbing shoe **104**, whereby a current is passed through the gel-like material **124** applied to the surface **120**, thereby removing contaminants from the surface **120**.



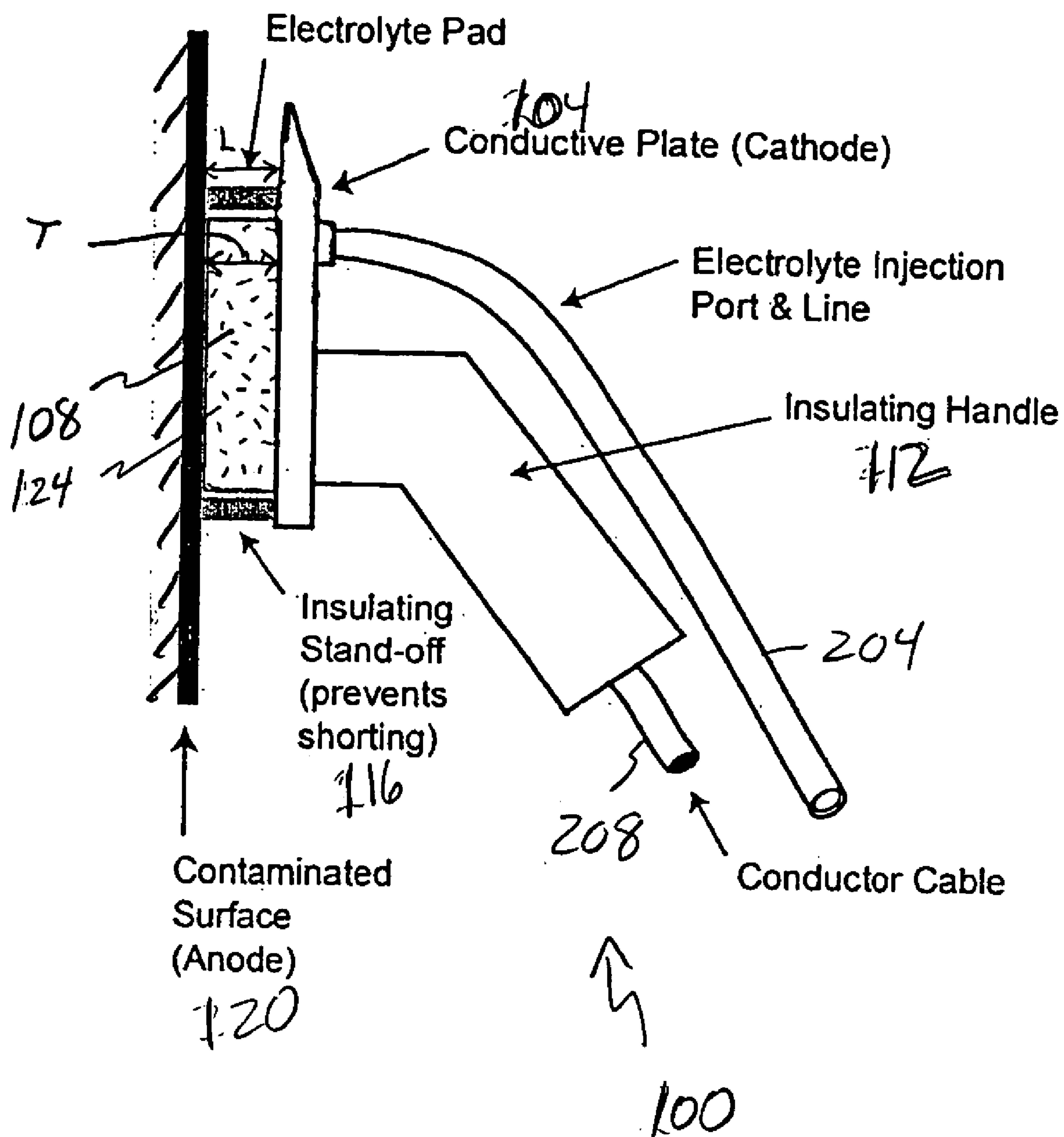


FIG. 1

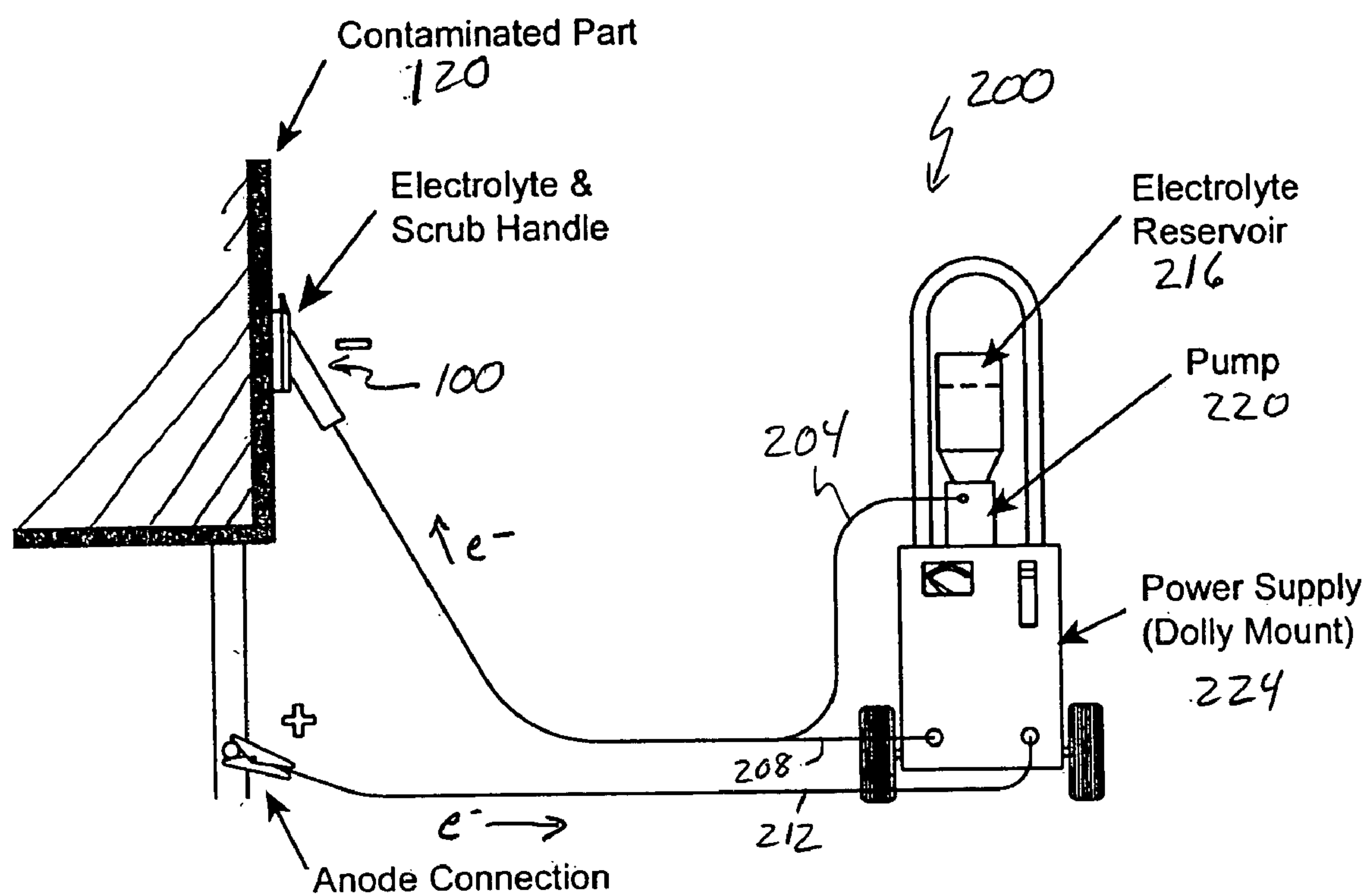
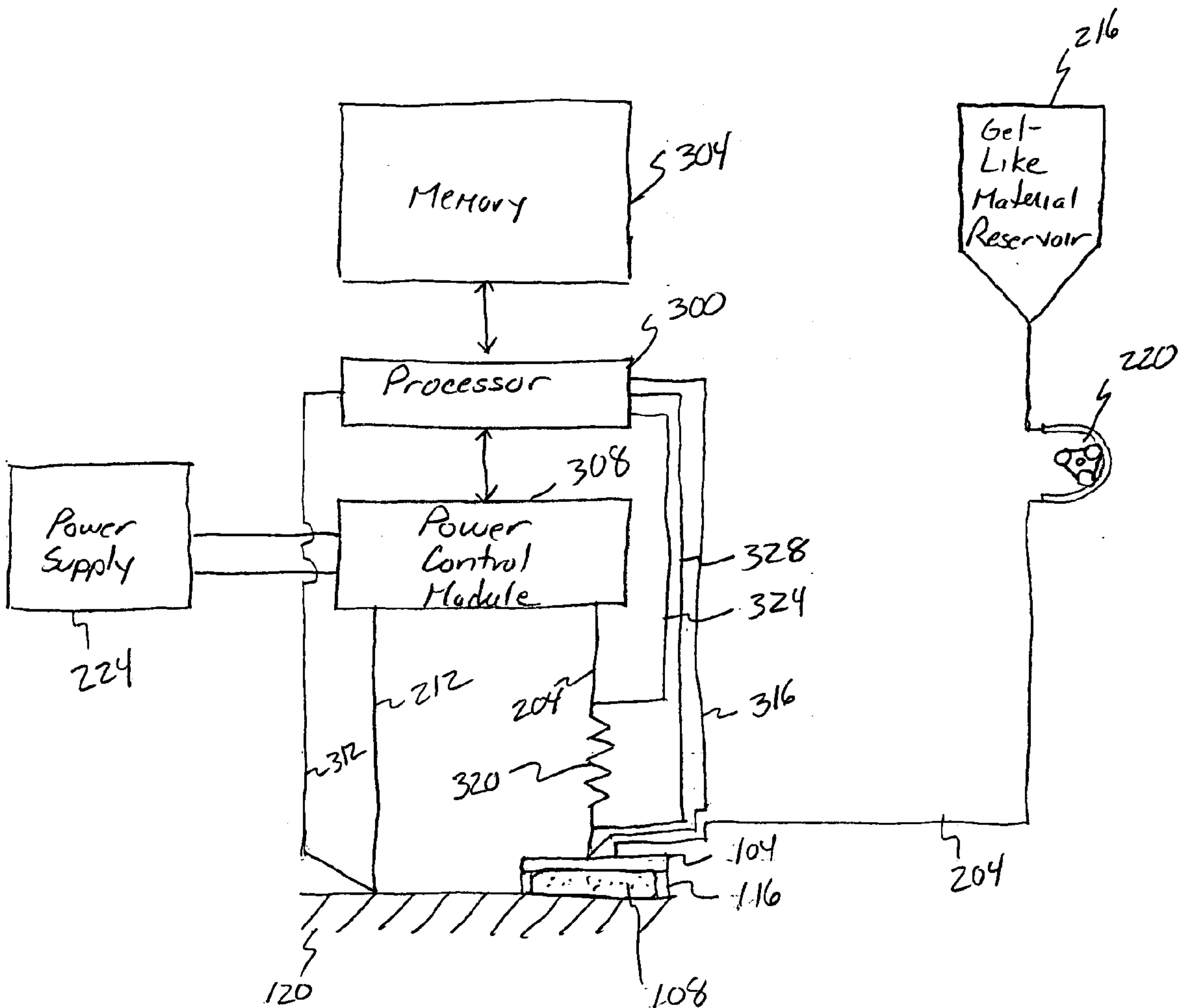


FIG. 2



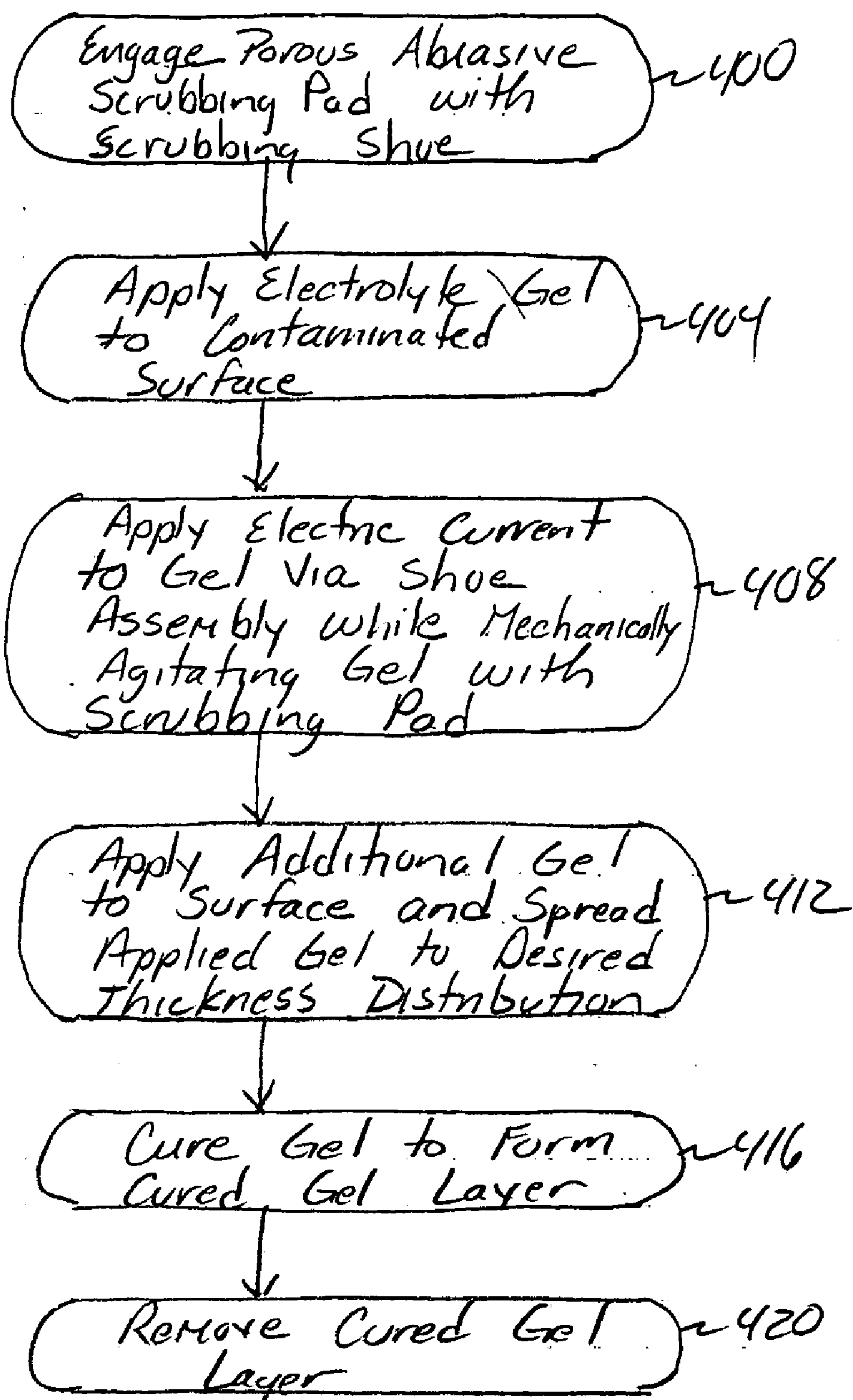
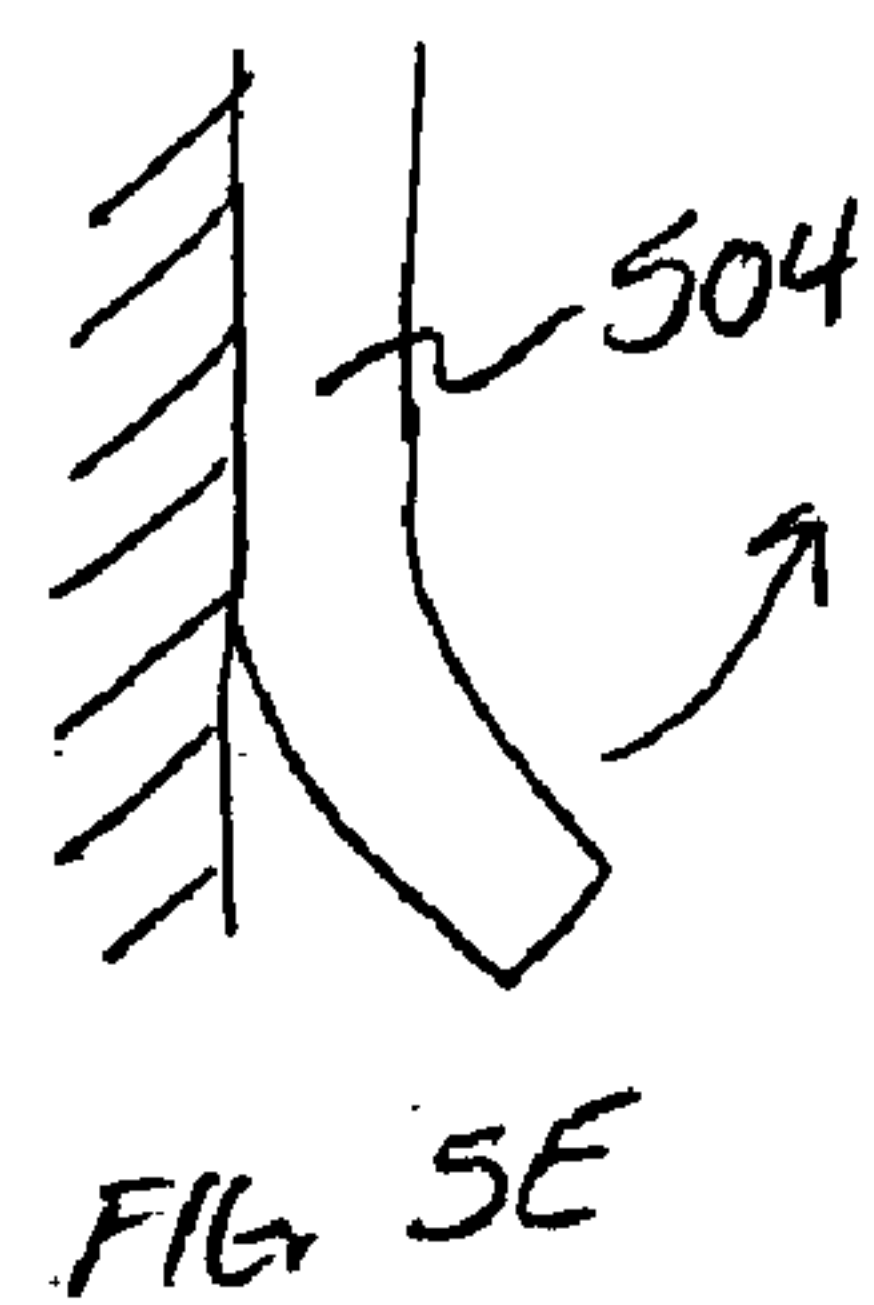
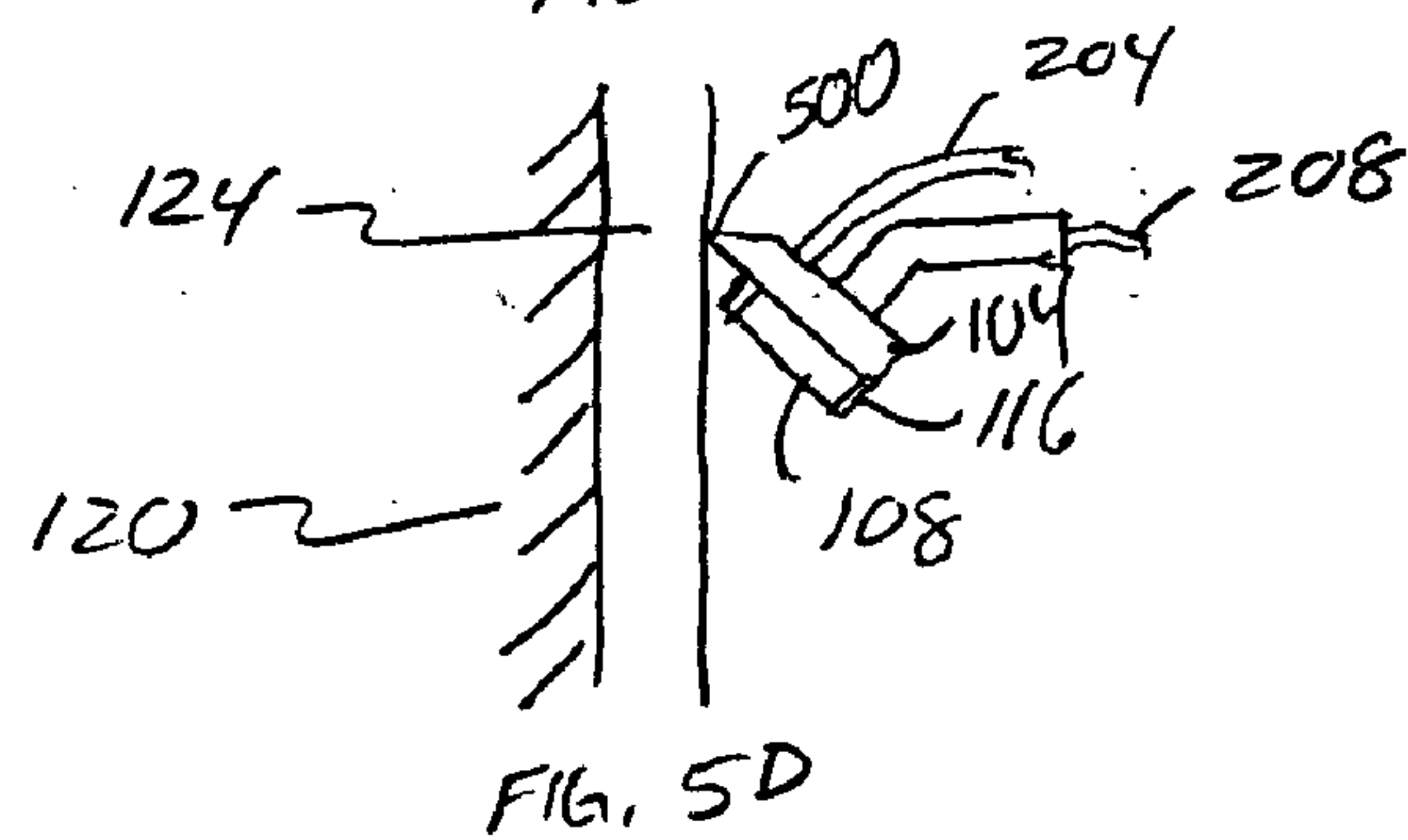
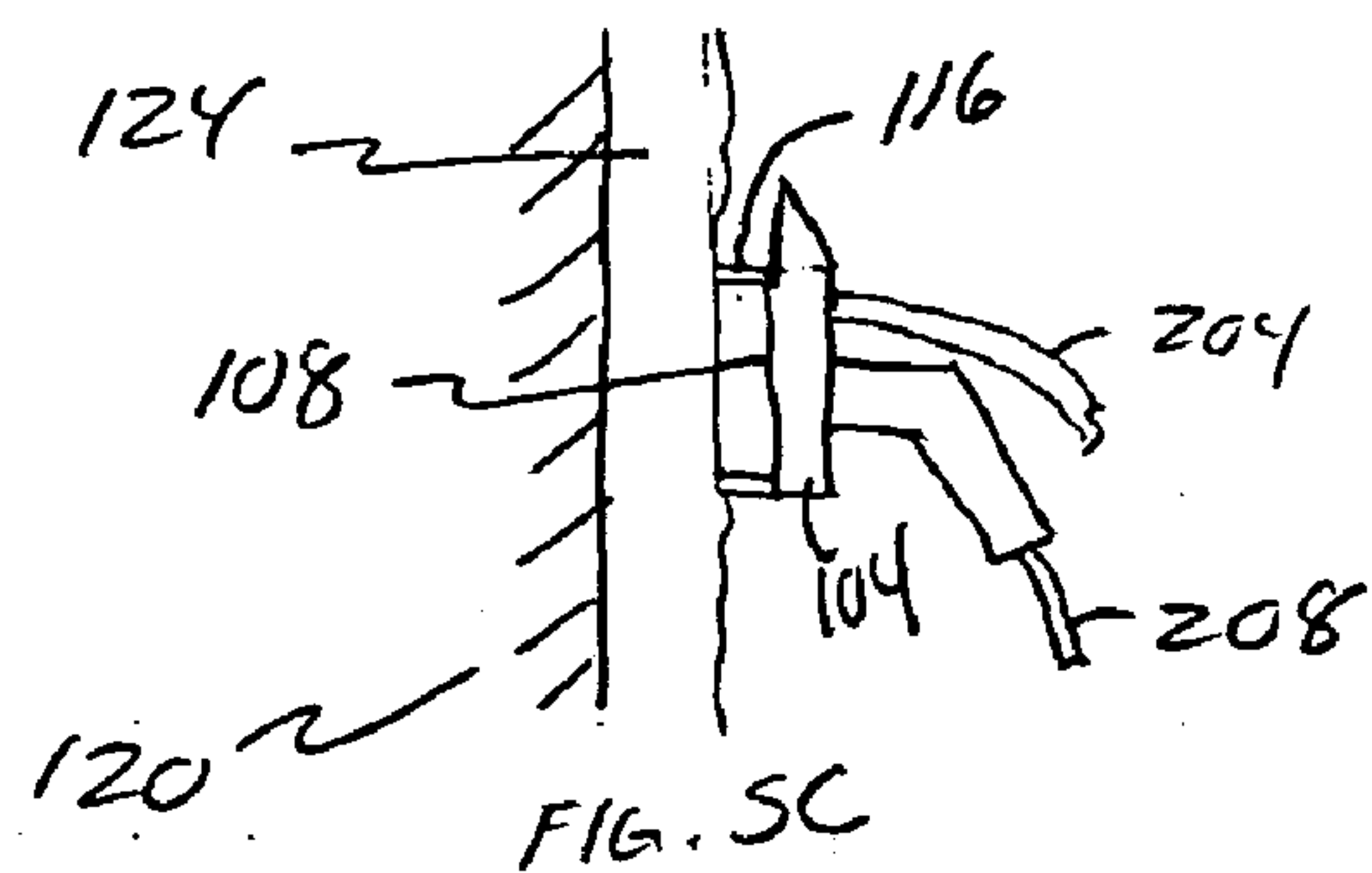
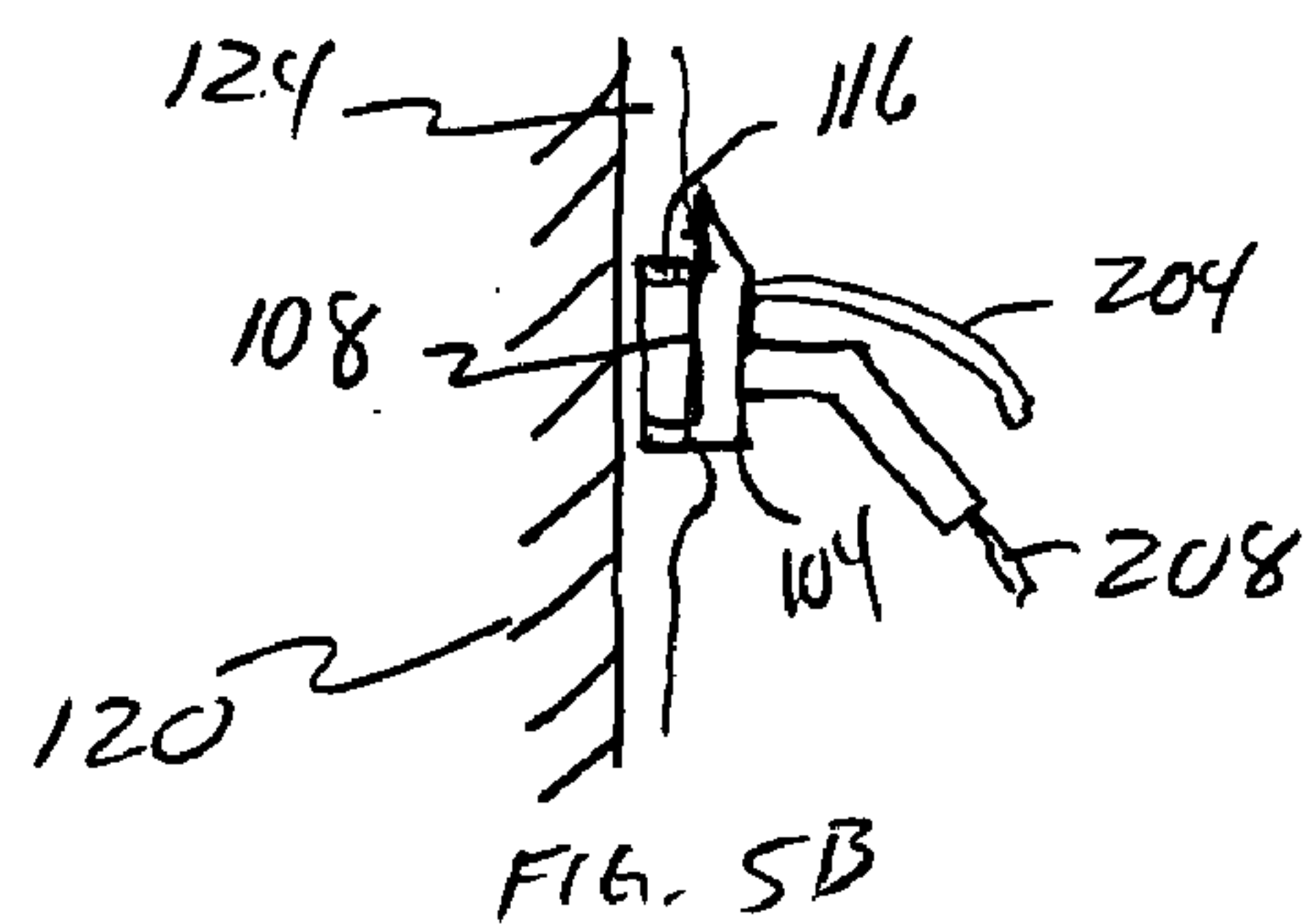
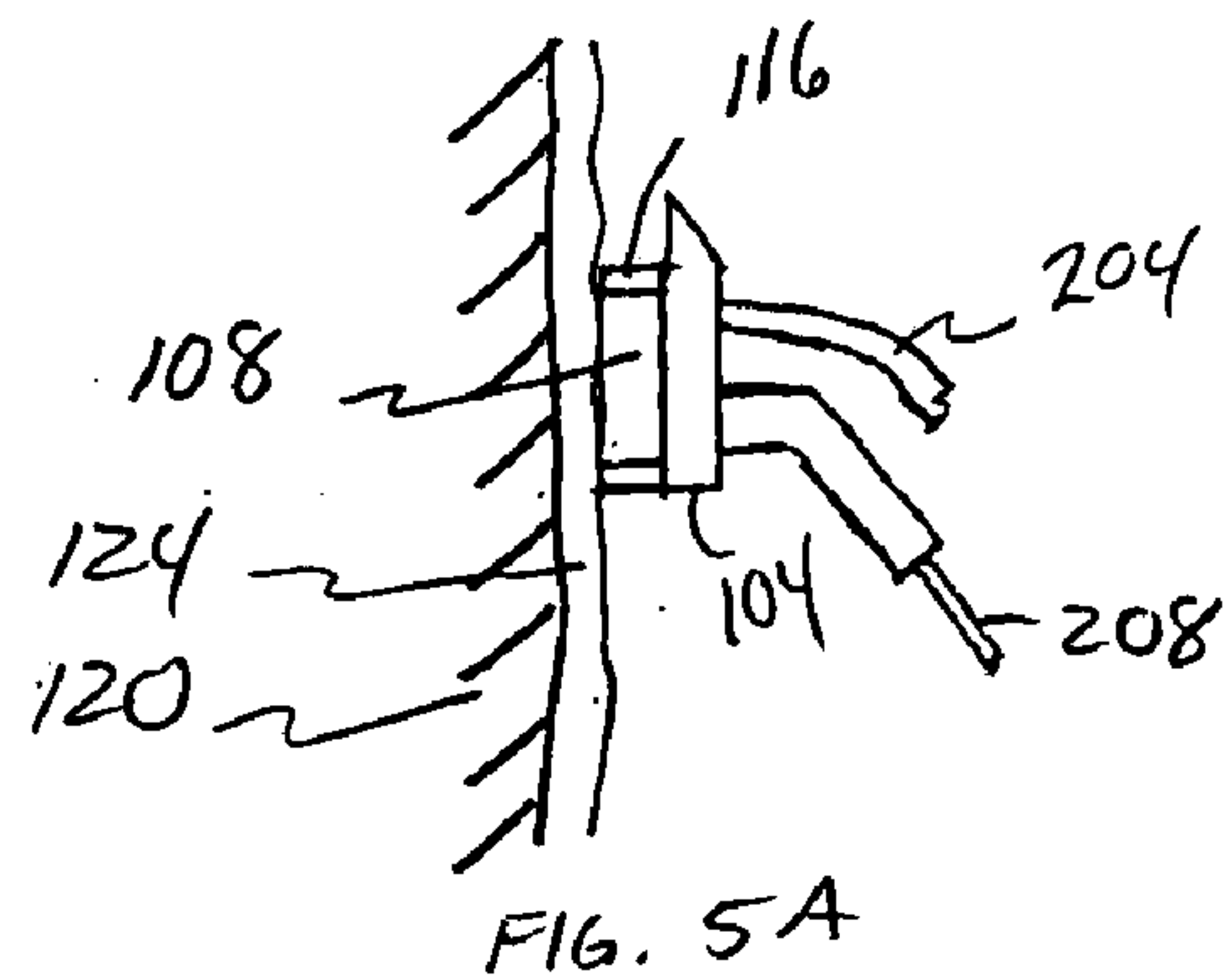


Fig. 4



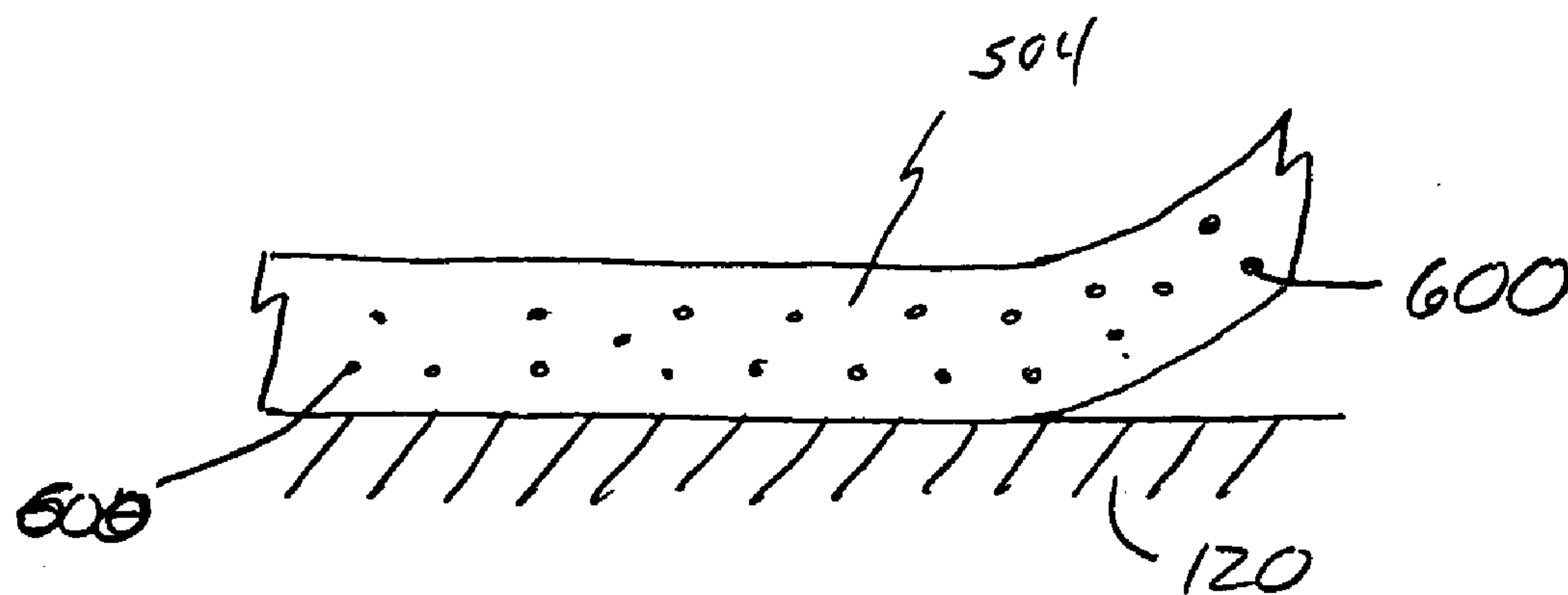


FIG. 6

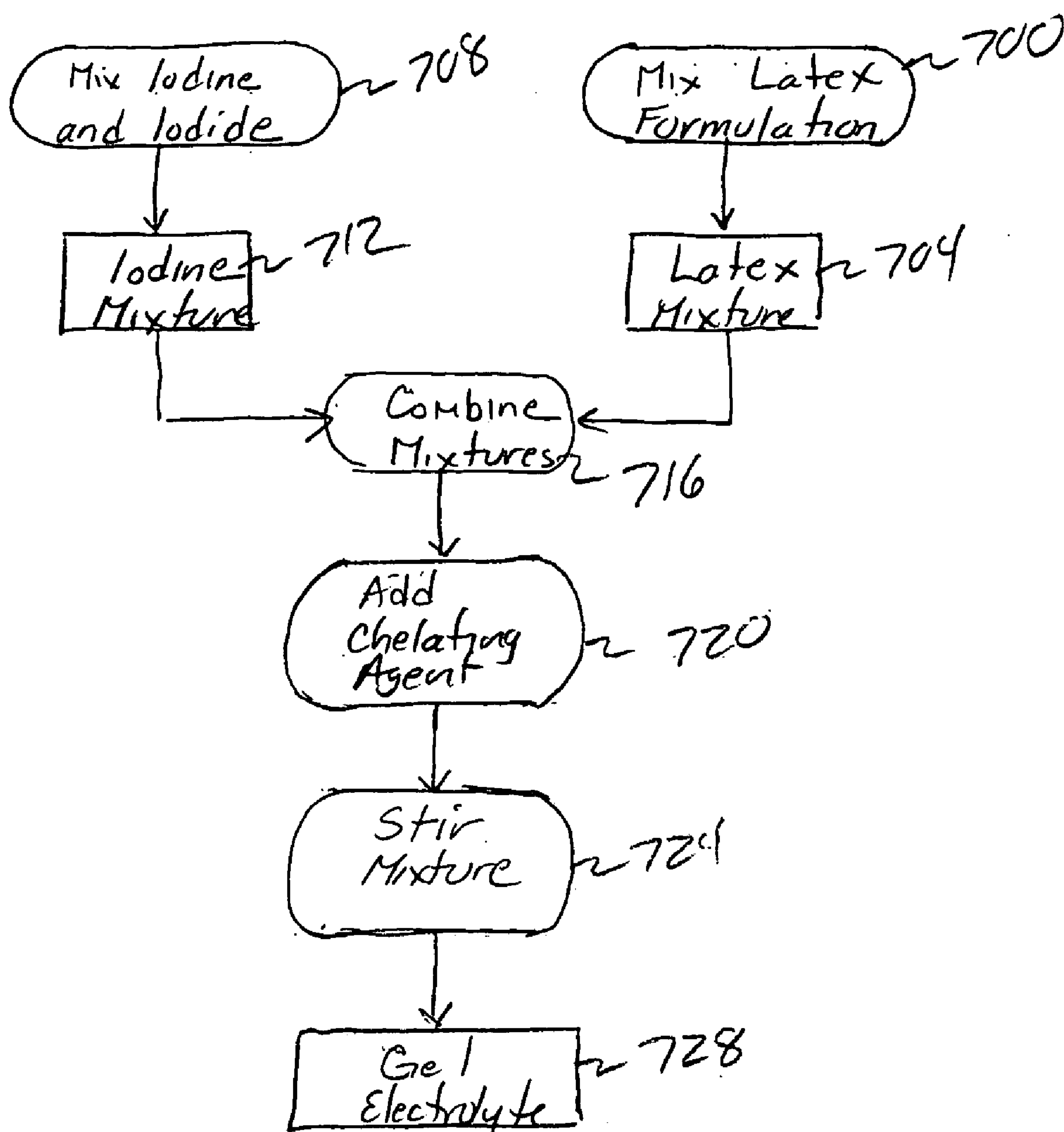


FIG. 7

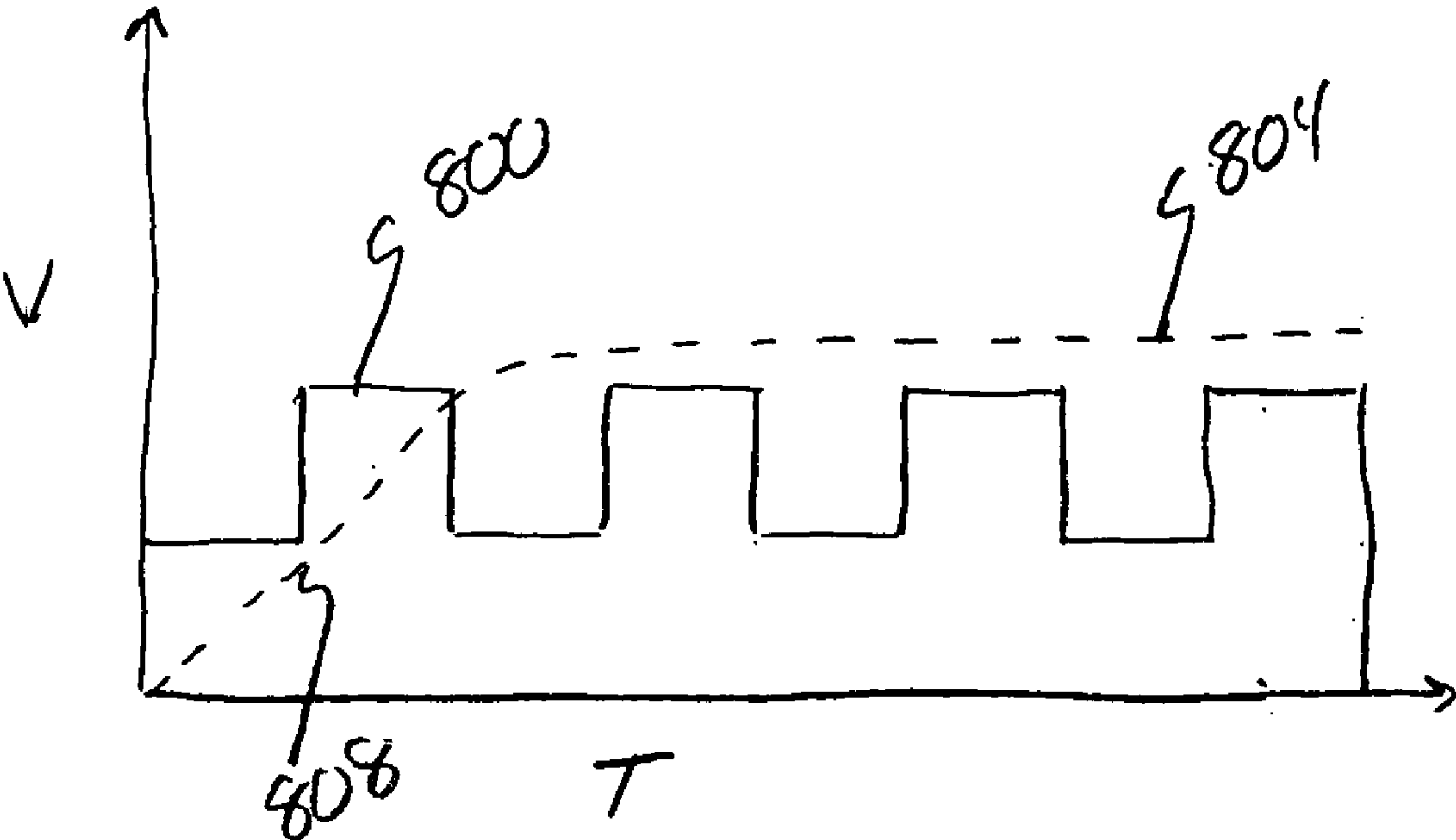


FIG. 8

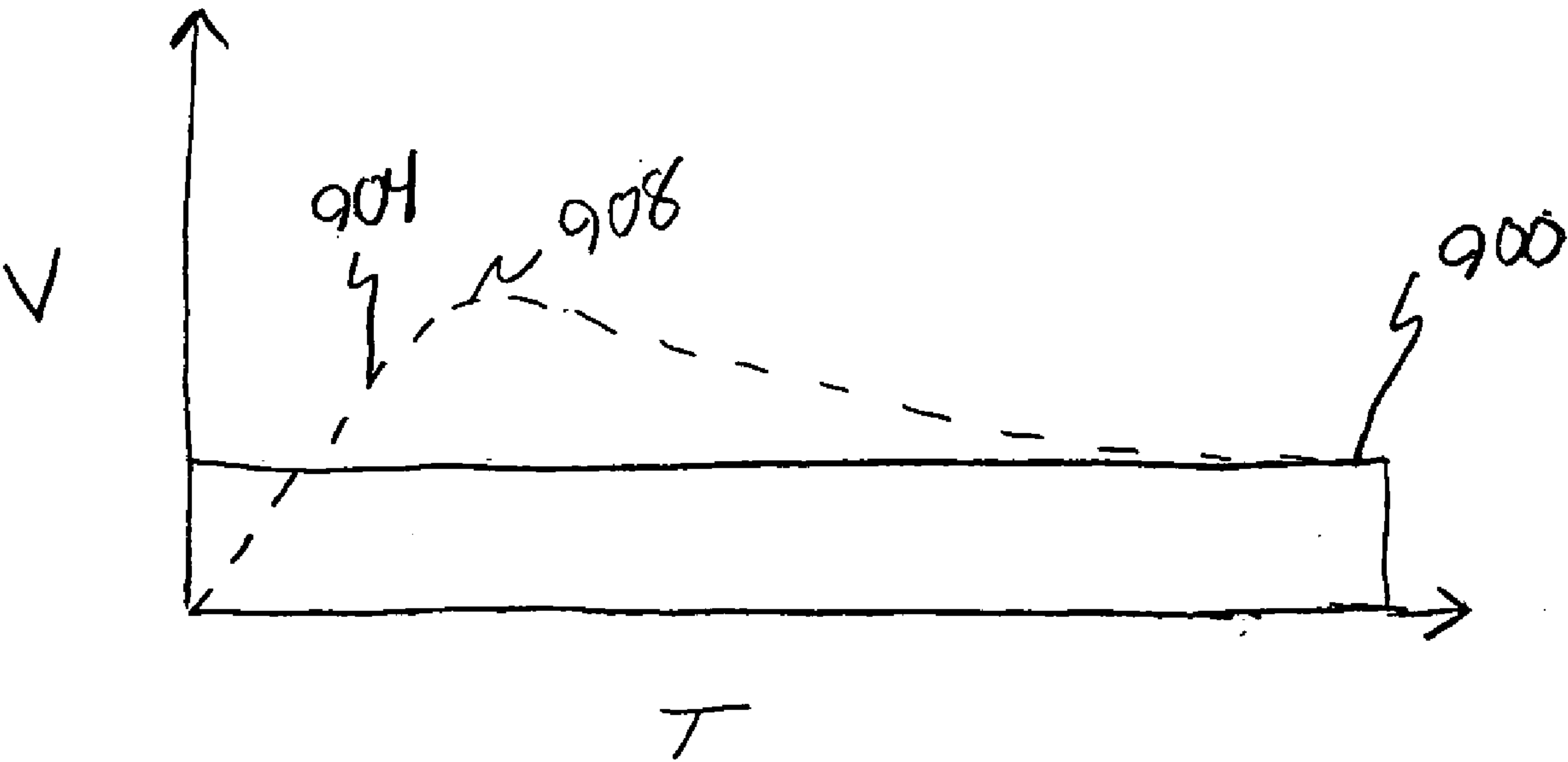


FIG. 9

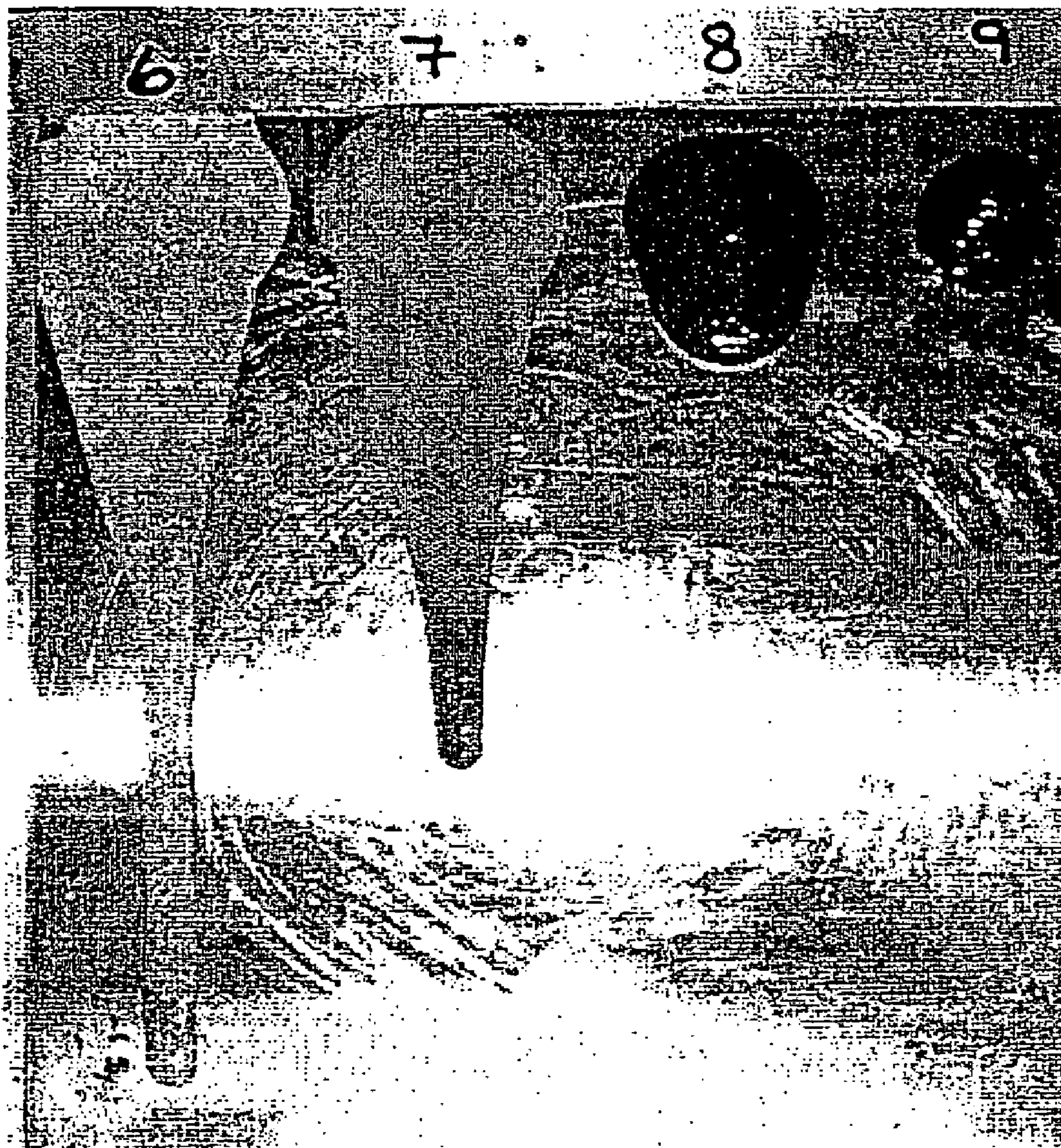


FIG. 10

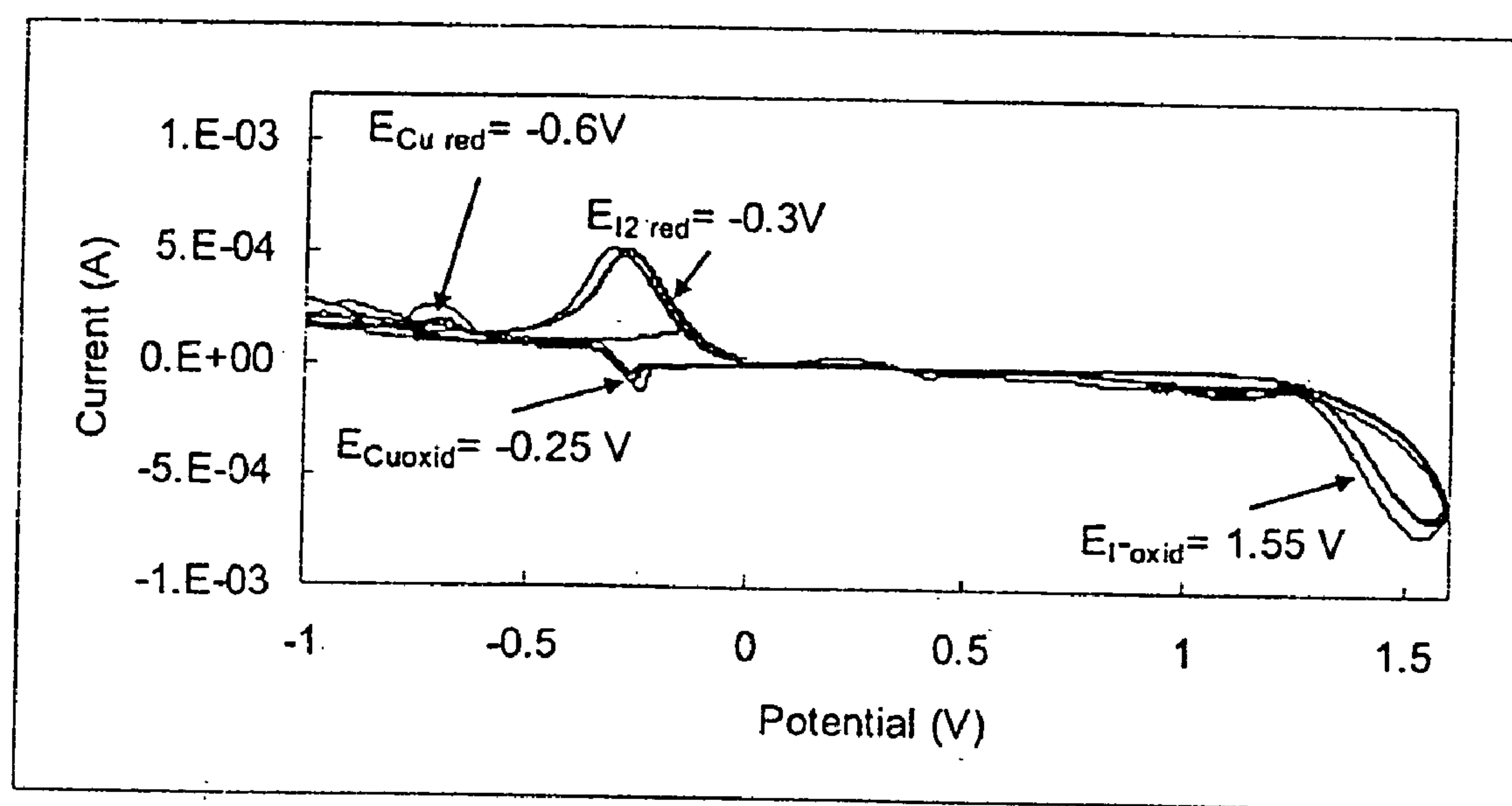


FIG. 11

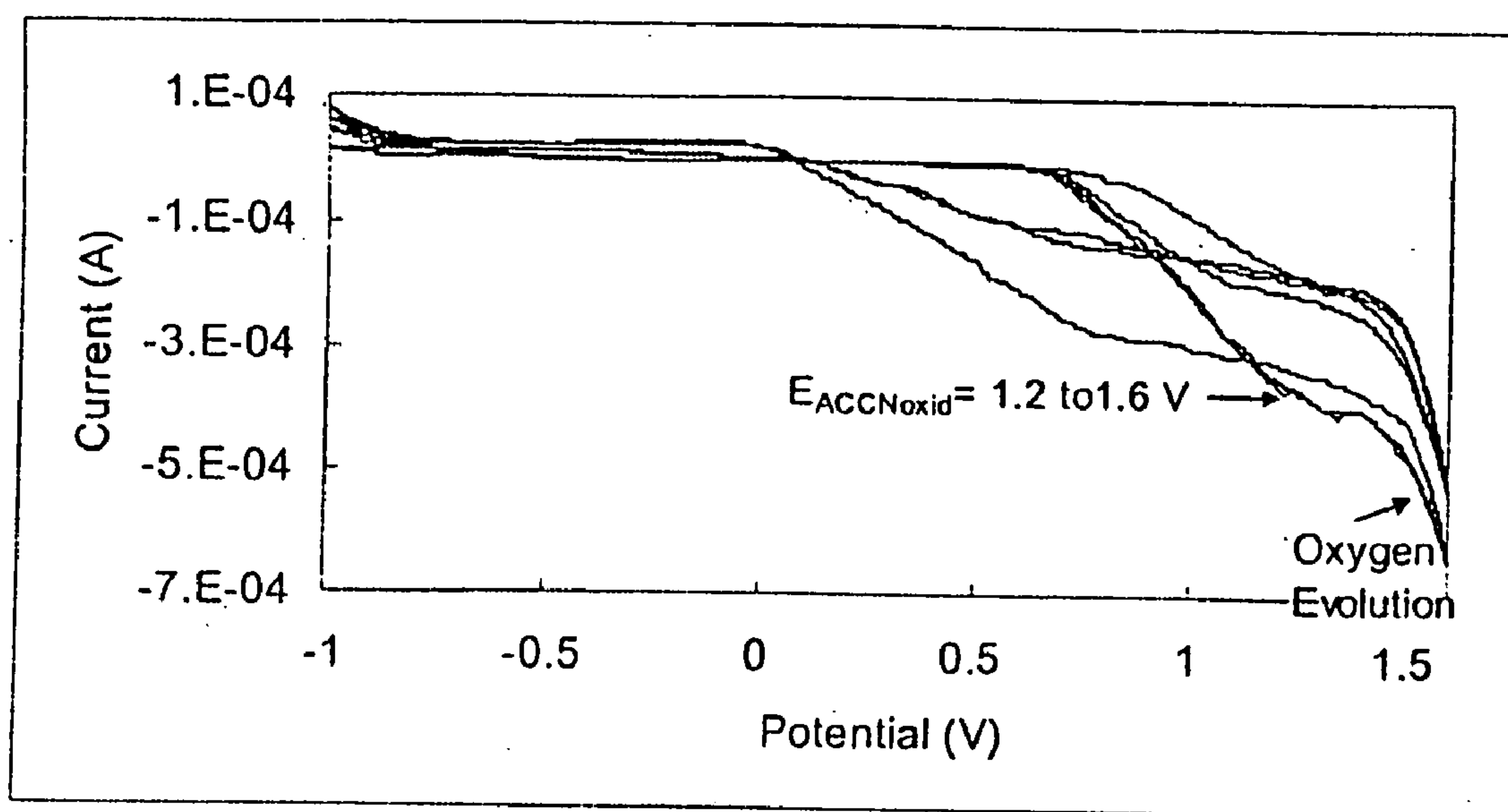


FIG. 12

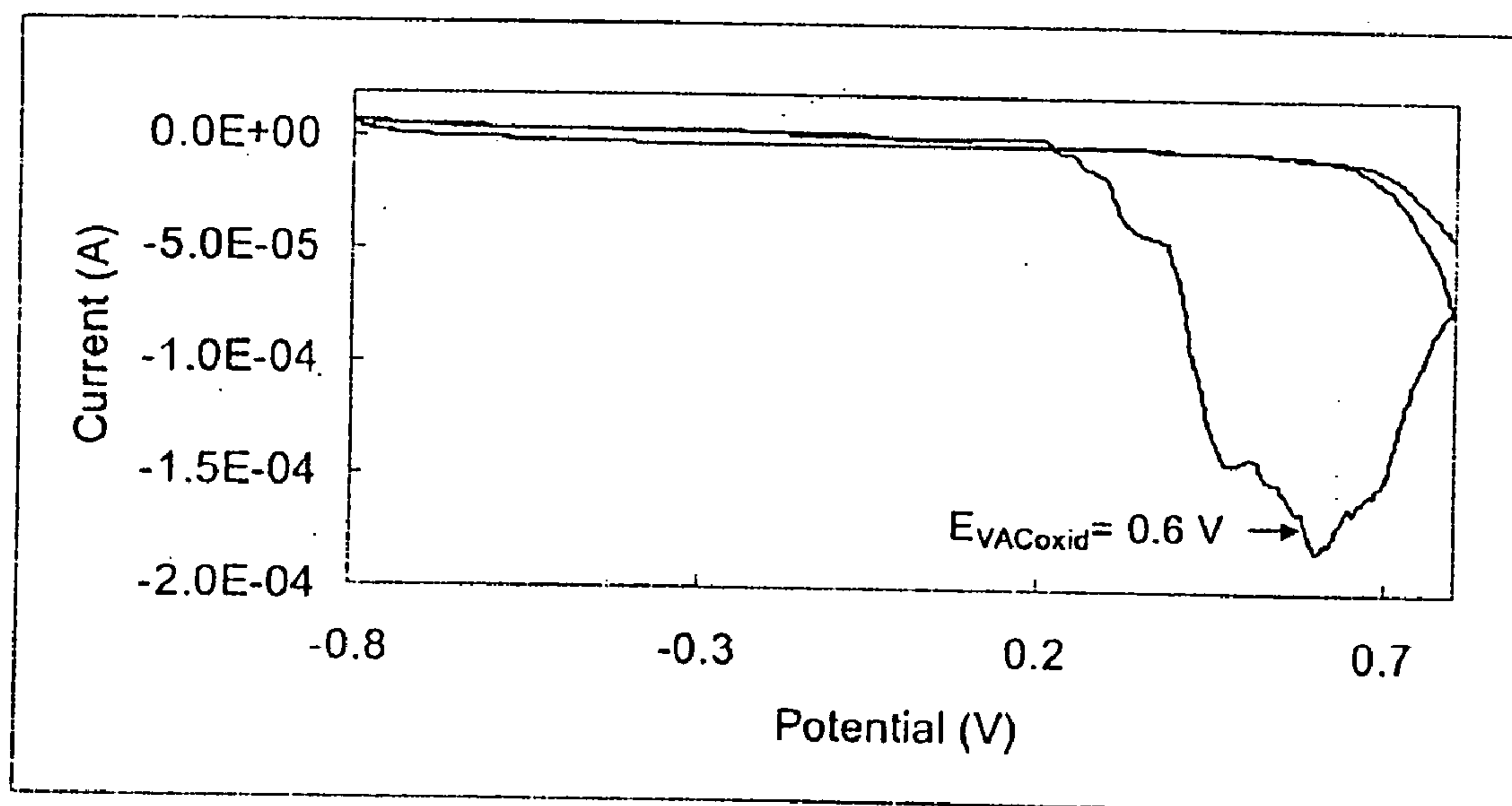


FIG. 13

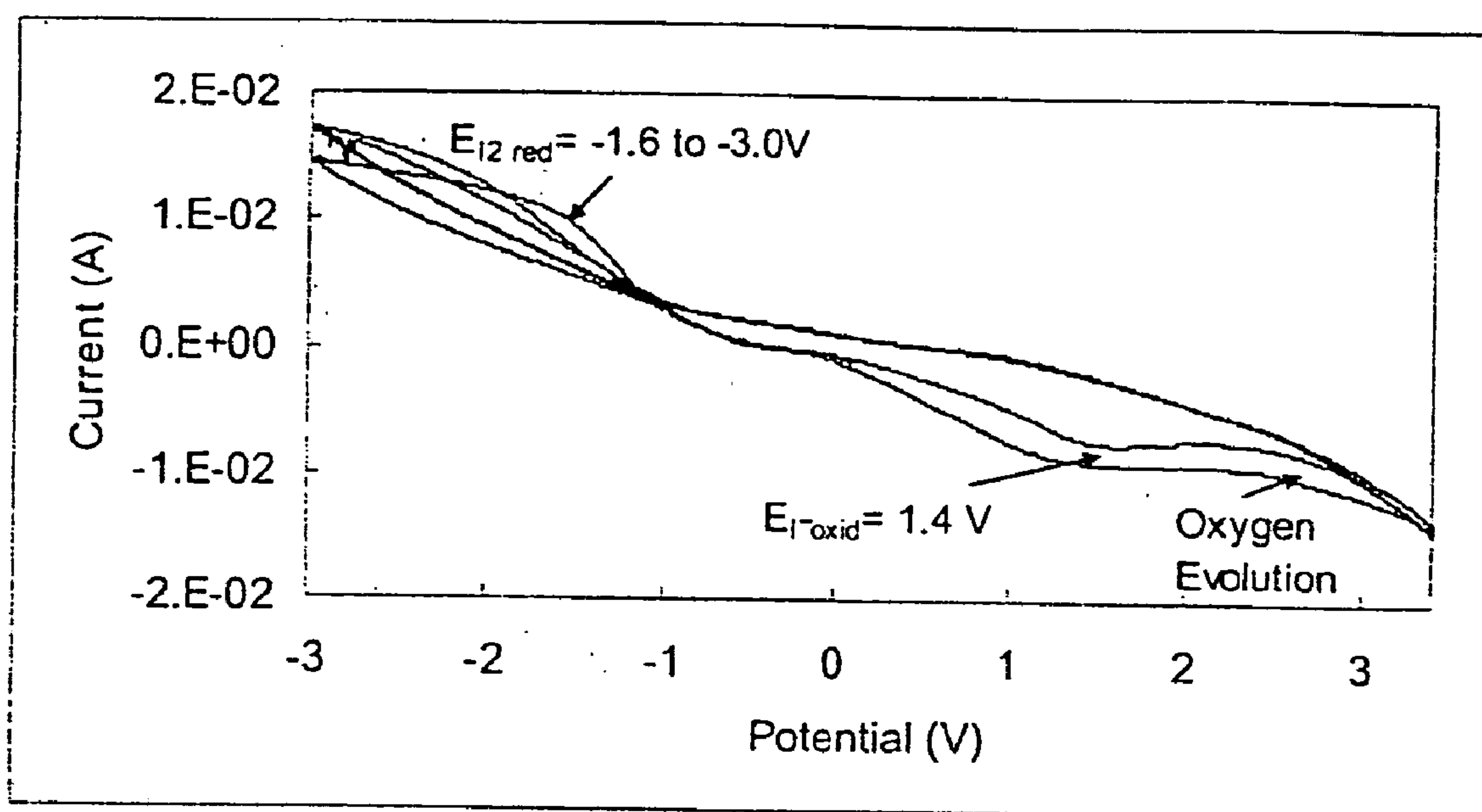


FIG. 14

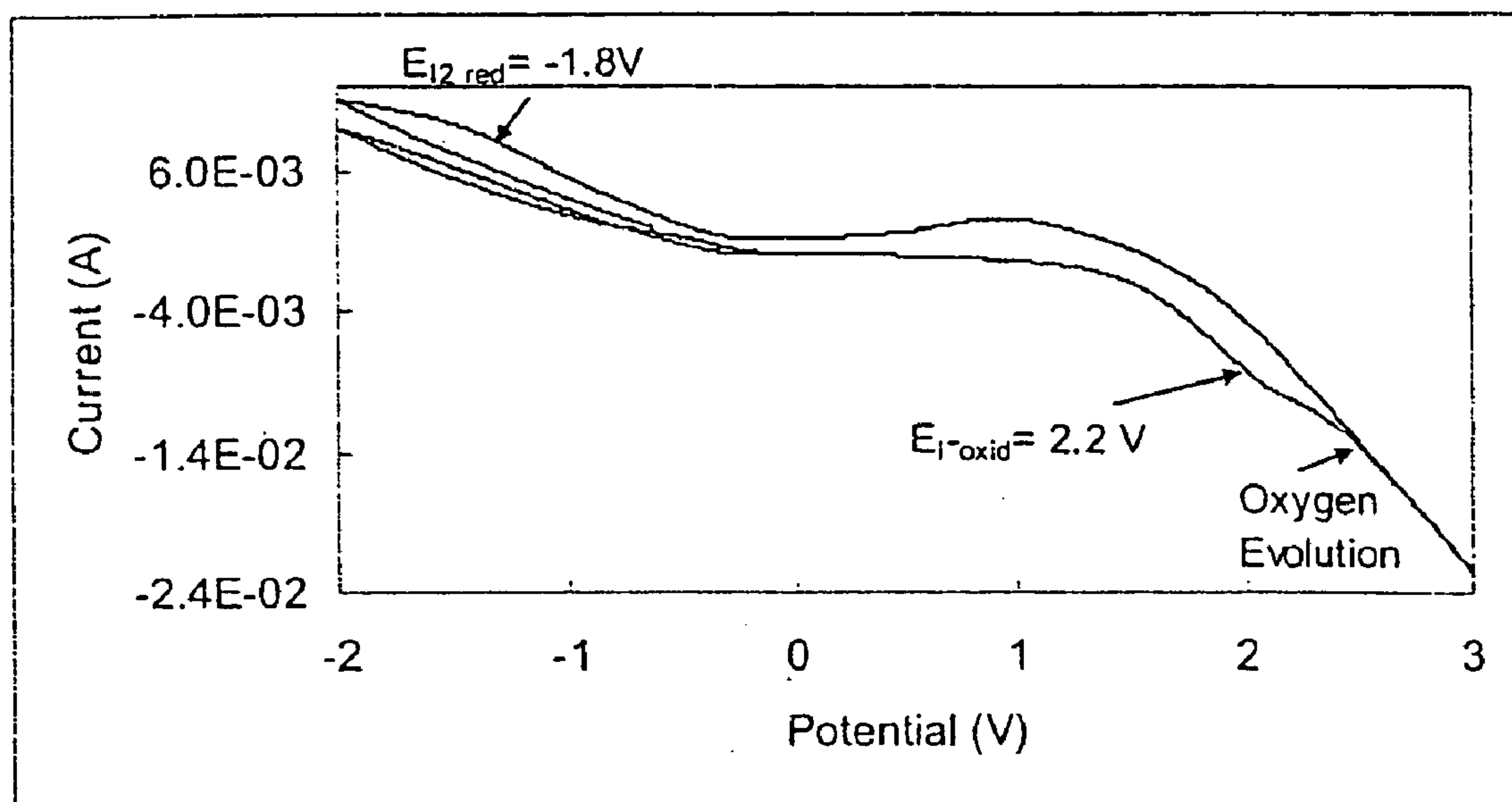


FIG. 15

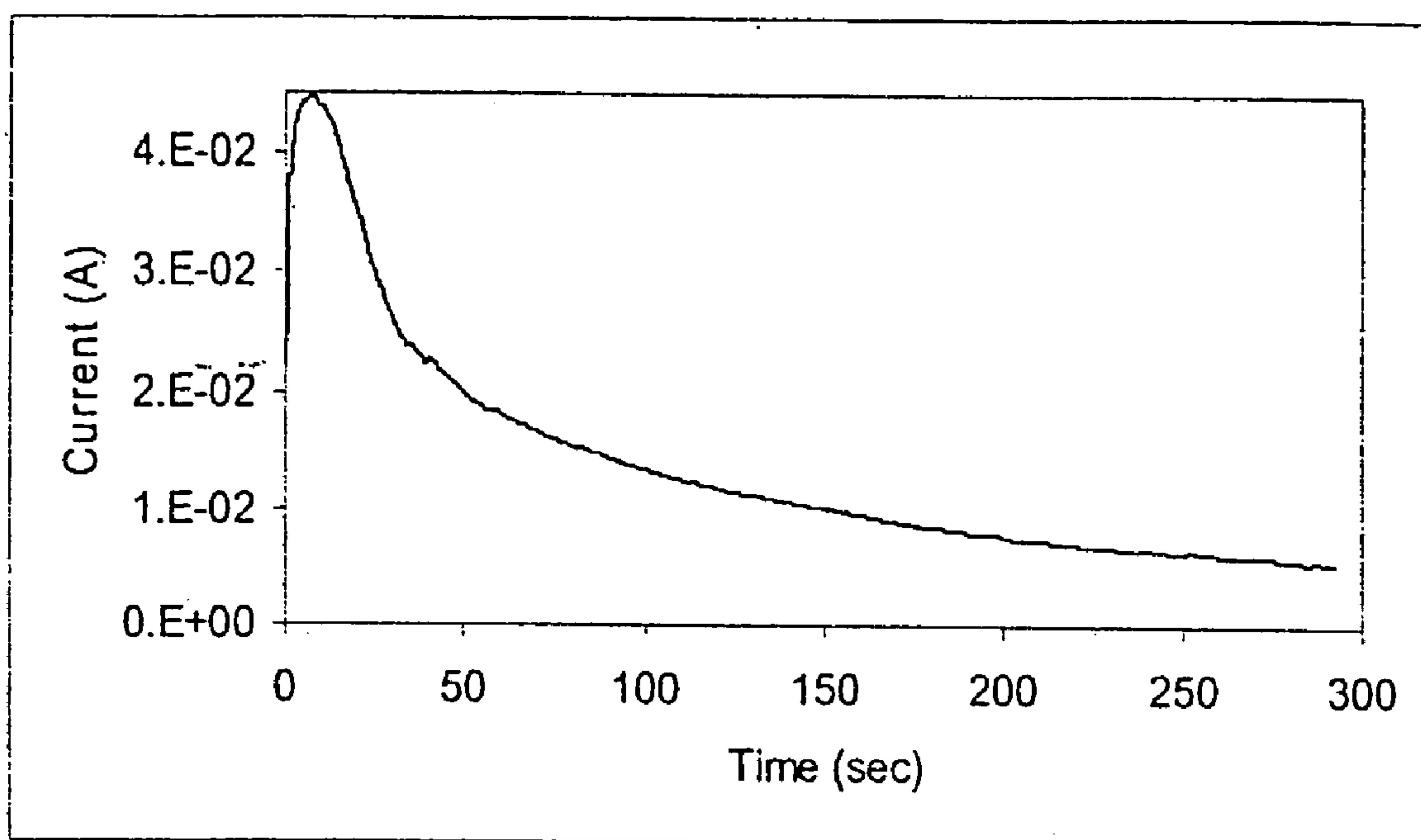


FIG. 16

Voltage Vs. Time for chronoamperometry waveform

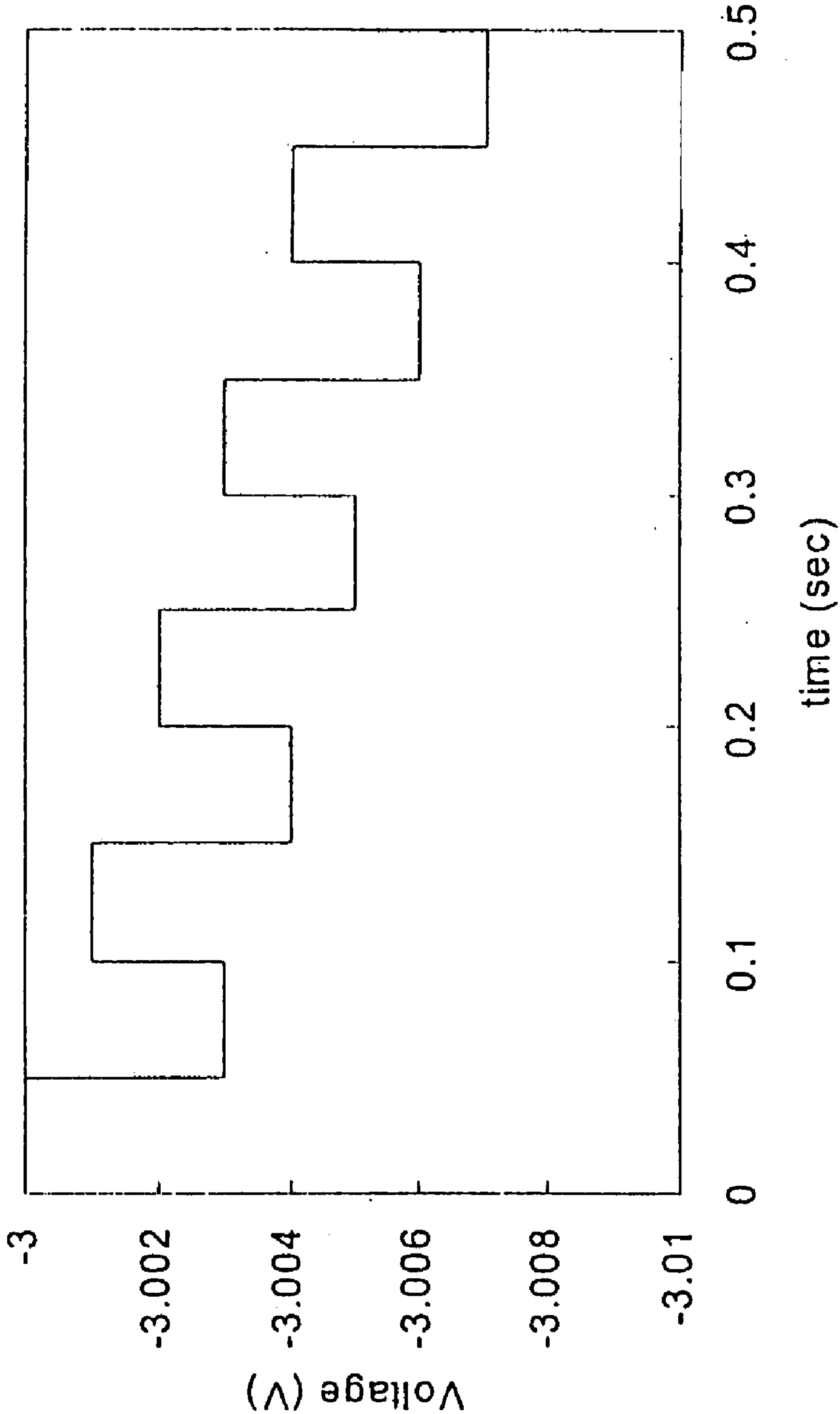


FIG. 17

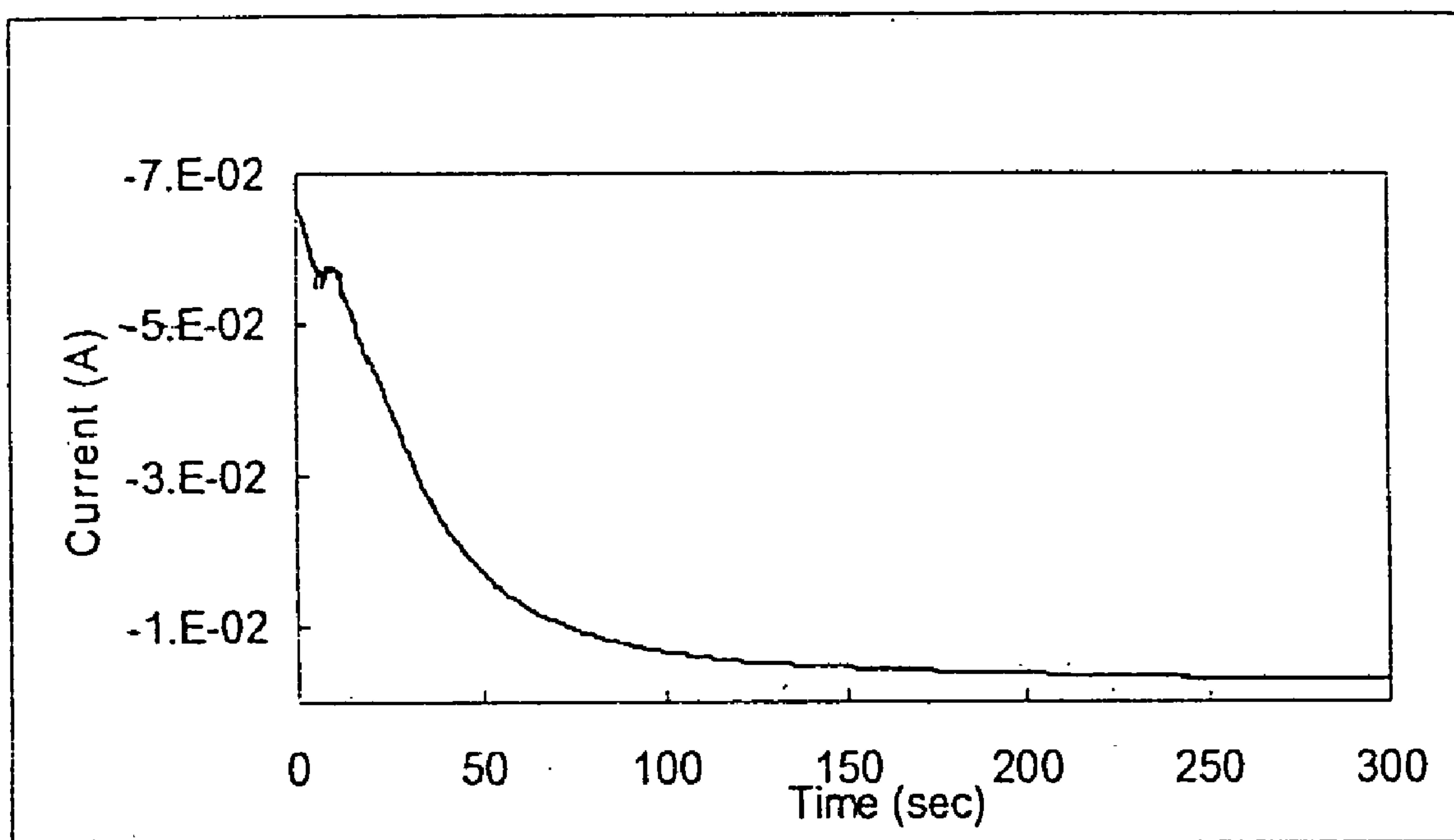


FIG. 18

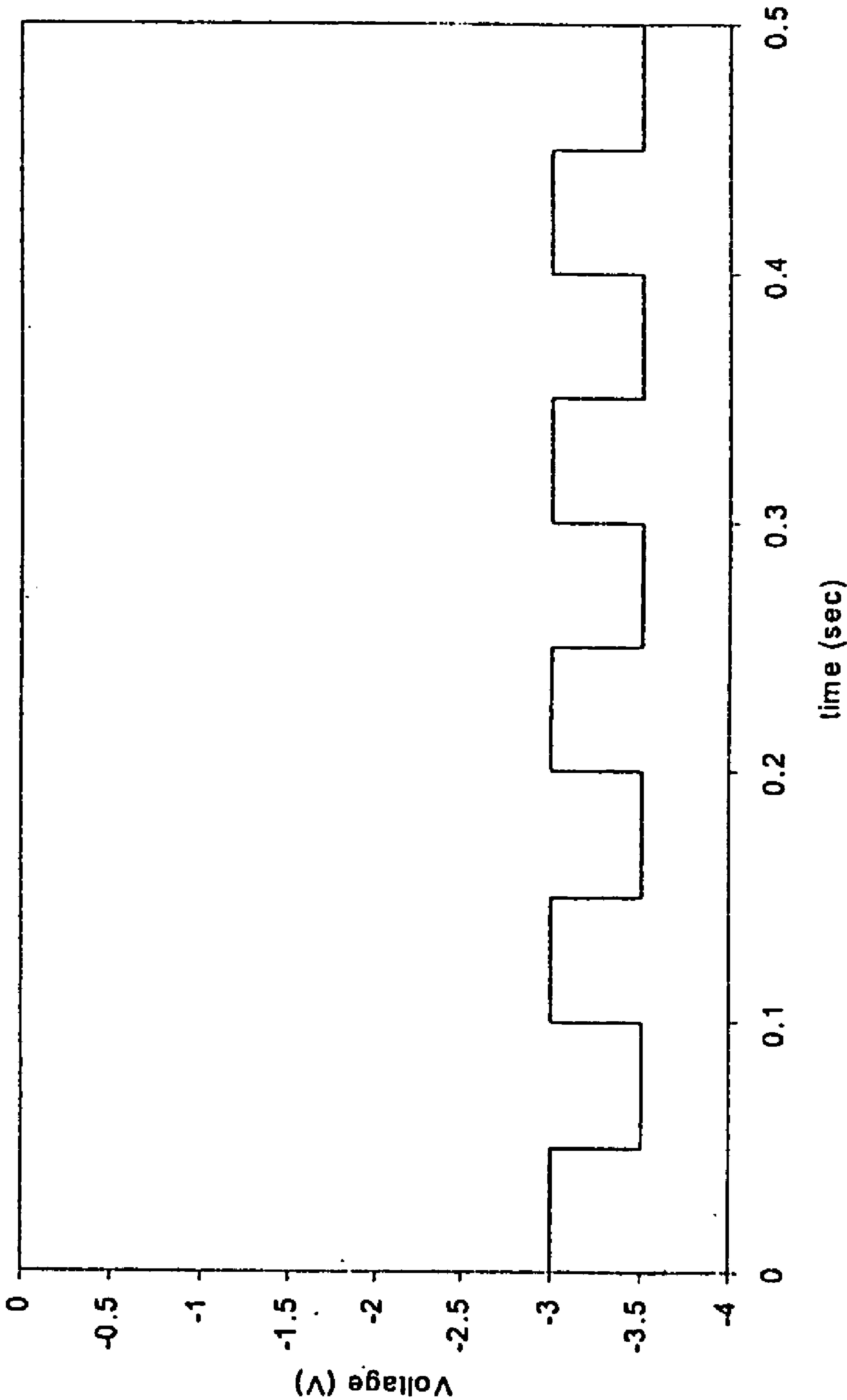


FIG. 19

ELECTRO-DECONTAMINATION OF CONTAMINATED SURFACES

CROSS REFERENCE TO RELATED APPLICATION

[0001] The present application claims the benefits of U.S. Provisional Application Serial No. 60/486,493, filed Jul. 10, 2003, entitled "Electrodecontamination for Mitigation of Airborne Contamination", which is incorporated herein by this reference.

STATEMENT REGARDING FEDERALLY SPONSORED RESEARCH OR DEVELOPMENT

[0002] The U.S. Government has a paid-up license in this invention and the right in limited circumstances to require the patent owner to license others on reasonable terms as provided for by the terms of Contract Nos. DE-FG03-00ER82934 and DE-FG02-03ER83591 awarded by the Department of Energy.

FIELD OF THE INVENTION

[0003] The invention relates generally to decontamination of objects and surfaces and particularly to electrolytic decontamination of radioactive contaminated surfaces.

BACKGROUND OF THE INVENTION

[0004] Radioactive contamination of objects can occur in a wide variety of applications, such as nuclear-powered utility power plants, submarines, and ships, nuclear weapons production plants and storage facilities, petrochemical plants and refineries, and uranium mines and refineries. In these applications, various types of equipment, including piping, vessels, pumps, valves, and the like, are exposed to surface contamination by man-made or naturally occurring radionuclides, such as radioactive isotopes of plutonium, uranium, cesium, cobalt, zirconium, strontium, radium, polonium, thorium, and daughter products of uranium and plutonium.

[0005] Surface contamination by radioactive materials is normally of two types. One type of surface contamination is known as smearable contamination. In smearable contamination, the radioactive materials on the surface can be removed by hand wiping the surface with a non-abrasive pad. Another type of surface contamination is known as fixed contamination. In fixed contamination, the radioactive materials have penetrated into microcracks, microcrevices, and other surface defects and irregularities and/or are chemically bonded to the surface. As a result, the materials are not removed simply by hand wiping the surface with a non-abrasive pad.

[0006] Currently available decontamination methods can be classified broadly under two categories: mechanical and chemical. Mechanical methods typically require surface abrasion to remove radioactive materials. Chemical methods, in contrast, typically require a chemical reaction to remove the radioactive materials from the surface.

[0007] Commonly used mechanical decontamination methods include vacuum cleaning, hydroblasting, sandblasting, blasting with other abrasives, flame cleaning, scraping, and scabbling. Mechanical decontamination methods can have drawbacks. Although they can remove both fixed and smearable contaminants, removed radioactive material is typically dispersed into the surrounding atmosphere, not only presenting a significant health hazard to decontamina-

tion personnel but also spreading to and contaminating other areas. Many of the mechanical methods are labor intensive, increasing both the cost of decontamination and decontamination personnel exposure time to radioactive materials. The complexity of many surface contours and shapes often renders decontamination by mechanical means difficult or impractical.

[0008] Commonly used chemical decontamination methods include water washing, steam cleaning, scrubbing with detergents, acids, caustics, and solvents, electro-chemical stripping, and strippable coatings. The water washing, steam cleaning, and scrubbing techniques can have drawbacks. They can require long treatment times to adequately decontaminate a surface because of low ion exchange rates and the need to apply chemical solutions at elevated temperatures, increasing the complexity and cost of the decontamination system.

[0009] Electro-chemical stripping and strippable coatings can overcome some of these drawbacks. In electrochemical stripping, the contaminated surface (which is conductive) is configured as an anode or cathode while an opposing electrode is configured as the cathode or the anode, respectively. An electrolyte solution is applied to the surface and the surface subjected to a voltage and therefore electrical current. Surface contaminants are electrolytically stripped from the surface into the electrolyte. Although electro-chemical stripping works well for objects that are fully immersed in an electrolyte bath, it does not work well for stationary surfaces or objects too large for full immersion in the bath. When applied to a surface, the liquid electrolyte, under the force of gravity, typically runs down the surface and becomes an uncontrolled carrier of the radioactive materials to other locations. Moreover, electrolytes can endanger personnel through their use of hazardous chemicals. Strippable coatings, in contrast, are viscous chemical formulations that are applied to the contaminated surface and capture the contaminants. After curing, the coating may be removed by stripping the coating off of the surface. Although strippable coatings are effective for smearable contaminants, they can have a very low removal rates for fixed contaminants.

SUMMARY OF THE INVENTION

[0010] These and other needs are addressed by the various embodiments and configurations of the present invention. The present invention removes contaminants from contaminated surfaces using a combination of electro-chemical and strippable coating techniques.

[0011] In one embodiment, a method for treating (e.g., decontaminating) an object is provided. The method includes the steps of:

[0012] (a) applying a gel-like material to a contaminated surface of the object;

[0013] (b) passing an electrical current through the applied gel-like material to drive the contaminants into the applied gel material.

[0014] (c) curing the gel-like material to form a cured, strippable layer of the material; and

[0015] (d) removing the cured layer from the object. As used herein a "gel-like material" refers to a viscous and/or thixotropic material commonly having a semi-solid or gel consistency. It may be in the form of a colloid in which a disperse phase is

combined with a continuous phase and only have the semi-solid or gel consistency when undisturbed but 20 flowing readily when manipulated or stirred or shaken.

[0016] The gel-like material, in one formulation, includes (a) an electrolytic agent, (b) a latex formulation, and (c) a chelating agent. A "latex" formulation refers to an aqueous suspension of a natural or synthetic polymer, which is typically a hydrocarbon. A "chelating agent" refers to a type of coordination compound in which a central metal cation, such as Co^{2+} , Ni^{2+} , Cu^{2+} , or Zn^{2+} , is attached by coordinate links to two or more nonmetal atoms in the same molecule, called ligands. Heterocyclic rings are typically formed with the central (metal) atom as part of each ring. Ligands offering two groups for attachment to the metal are termed bidentate, three groups, tridentate, etc.

[0017] The electrolytic agent can be any acid, salt or base. Preferably, it is a metal and/or metal-containing compound with the metal preferably being selected from Groups 6-12 and 17 of the Periodic Table of the Elements and even more preferably being a halogen or transition metal. Iron and iodine-containing compounds are even more preferred and iodine and iodine-containing compounds most preferred.

[0018] The latex formulation typically includes one or more monomers, an initiator, a surfactant, and water. An "initiator" refers to an agent used to start the polymerization of a monomer or oligomer. Examples of initiators include organic and inorganic peroxides. A particularly preferred latex formulation includes vinyl and acrylic monomers.

[0019] The chelating agent can be any suitable material that can form a complex with a selected metal. Examples of suitable chelating agents include ethylene-diaminetetraacetic acid (EDTA), nitrilotriacetic acid ($\text{N}(\text{CH}_2\text{COOH})_3$), ethyleneglycol-bis-(β -aminoethyl ether)-N,N-tetraacetic acid ($\text{HOOCCH}_2)_2\text{NCH}_2\text{CH}_2\text{OCH}_2\text{CH}_2\text{OCH}_2\text{CH}_2\text{NN}(\text{CH}_2\text{COOH})_2$, ethylenediamine, porphine, and dimercaprol.

[0020] While not wishing to be bound by any theory, it is believed that, when voltage is applied to the gel-like material via the decontaminating apparatus, electrochemical reactions, optimally driven by a power waveform supplied by the power source, strip away the contaminated surface's top-most layers of atoms along with other contaminants that may be present. Current is carried by water molecules and the electrolytic agent in the gel-like material, and the electrolytic agent contributes electrons to oxidation reactions occurring at the anode and receives electrons from the reduction reactions occurring at the cathode. Depending on the system configuration, anionic or elemental contaminants may also be oxidized to a cation, and cation or elemental contaminants reduced to an anion. As a result, the material is stripped away, and becomes entrained in the gel-like material. Gentle scrubbing action using the abrasive pad on the contaminated surface disrupts oil and grease deposits, affording better electrical contact while simultaneously stirring or agitating the gel-like material to bring fresh gel-like material into contact with the contaminated surface. The decontaminating system can have a control to permit either reduction or oxidation potentials to be applied to the contaminated surface so that both metals and metal oxides, respectively, can be removed successfully. This capability can allow the system to remove effectively oxidized metal layers (e.g., rust) that may be present on the contaminated surface.

[0021] The present invention can have a number of advantages. For example, the present invention can quickly remove high levels of both fixed and smearable contaminants while minimizing airborne entrainment of the contaminants. The gel-like material captures and entrains the contaminants as they are removed from the surface. It is not be labor intensive and controls decontamination personnel exposure to radioactive materials. It can treat the surfaces in relatively short time periods and work well for large stationary surfaces, regardless of orientation due to the gel-like consistency of the material. It can use nontoxic and non-hazardous components, which can not only provide higher levels of personnel safety but also result in substantial cost savings. It achieves a magenta color when applied and a yellow color when cured, which are consistent with color codings for radioactively contaminated materials. The gel-like material can be left in place as a protective barrier to prevent recontamination of the surface. When removed, the cured gel-like material contains the radioactive materials and is in a form, namely radioactively contaminated plastics, that is already handled and familiar to current decontamination operations personnel. The decontamination system of the present invention can be compact, portable, easy to deploy, and placed in hot storage when not in use. It can minimize or eliminate entirely dangerous off-gases, making it suitable for use in tight quarters and confined spaces with limited egress, such as under gloveboxes, inside tanks and vessels, or in overhead ceiling spaces and pipe chases.

[0022] These and other advantages will be apparent from the disclosure of the invention(s) contained herein.

[0023] The above-described embodiments and configurations are neither complete nor exhaustive. As will be appreciated, other embodiments of the invention are possible utilizing, alone or in combination, one or more of the features set forth above or described in detail below.

BRIEF DESCRIPTION OF THE DRAWINGS

[0024] FIG. 1 is a side view of a decontaminating apparatus according to an embodiment of the present invention;

[0025] FIG. 2 is a side view of a decontaminating system according to an embodiment of the present invention;

[0026] FIG. 3 is a block diagram of the decontaminating system of FIG. 2;

[0027] FIG. 4 is a flowchart of the decontaminating process according to an embodiment of the present invention;

[0028] FIGS. 5A-E are various depictions of the steps in the decontaminating process of FIG. 4;

[0029] FIG. 6 depicts removal of the cured electrolytic gel-like coating according to an embodiment of the present invention;

[0030] FIG. 7 is a flowchart of a method of manufacturing the electrolytic gel-like material;

[0031] FIG. 8 is a voltage waveform according to an embodiment of the present invention;

[0032] FIG. 9 is a voltage waveform according to an embodiment of the present invention;

[0033] FIG. 10 shows the relative viscosities on a vertical surface for various gel-like material formulations;

[0034] FIG. 11 is a plot of current (Amperes) (vertical axis) against potential (Volts) (horizontal axis);

[0035] FIG. 12 is a plot of current (Amperes) (vertical axis) against potential (Volts) (horizontal axis);

[0036] FIG. 13 is a plot of current (Amperes) (vertical axis) against potential (Volts) (horizontal axis);

[0037] FIG. 14 is a plot of current (Amperes) (vertical axis) against potential (Volts) (horizontal axis);

[0038] FIG. 15 is a plot of current (Amperes) (vertical axis) against potential (Volts) (horizontal axis);

[0039] FIG. 16 is a plot of current (Amperes) (vertical axis) against time (Seconds) (horizontal axis);

[0040] FIG. 17 is a plot of current (Amperes) (vertical axis) against time (Seconds) (horizontal axis);

[0041] FIG. 18 is a plot of current (Amperes) (vertical axis) against time (Seconds) (horizontal axis); and

[0042] FIG. 19 is a plot of current (Amperes) (vertical axis) against time (Seconds) (horizontal axis).

DETAILED DESCRIPTION

The Decontamination System

[0043] The decontamination system effectively combines the underlying concepts of electro-chemical decontamination or electropolishing, strippable decontamination coatings, and mechanical decontamination. The system uses a viscous and/or thixotropic electrolytic gel-like material that is applied to the decontaminated surface, subjected typically to an applied voltage waveform (though a single voltage can alternatively be applied) causing electrolytic stripping of the fixed and smearable contaminants, and cured to form a strippable coating containing the electrolytically removed contaminants. Although the gel-like material is discussed with reference to removal of radioactive contaminants, it is to be understood that the gel-like material may be used for any contaminant or undesired material that will itself or that is deposited on a surface containing one or more substances that will themselves be oxidized or reduced in the presence of an applied voltage/electrical current.

[0044] FIGS. 1-2 depict a decontaminating system according to an embodiment of the present invention. The system 200 includes a decontaminating apparatus 100, gel-like material supply line 204, first and second conductor cables 208 and 212, gel-like material reservoir 216, pump 220, and power supply 224.

[0045] With reference to FIG. 1, the decontaminating apparatus 100 includes a scrubbing shoe 104 that receives a porous, permeable, and abrasive scrub pad 108, insulating handle 112, and insulating stand-off 116 (which prevents shorting and surrounds and receives the pad 108). The scrubbing shoe 104 acts as an electrode and is therefore composed of a conductive or semi-conductive material. The shoe 104 is electrically connected to the first conductor cable 208. The pad 108 is received and held in position by the stand-off 116. The pad is preferably formed of an insulating material, such as a polymeric material, impregnated with an inert abrasive material. The stand-off 116 has a thickness no more than that of the pad 108 to allow the pad 108 to contact the surface 120. To maintain desired current levels,

the standoff length ("L") typically ranges from about $\frac{3}{16}$ to about $\frac{3}{8}$ inches while the pad thickness ("T") typically ranges from about $\frac{1}{4}$ to about $\frac{1}{2}$ inches. Gel-like material 124 flows from the supply line 204, through (and saturates) the pores in the pad 108, to the surface 120 and establishes a conductive path from the conductive shoe 104 to the surface 120. A particularly preferred pad 108 is the Scotch-Brite™ pad manufactured by 3M™, which includes polymeric fibers impregnated with an abrasive material. In normal use the pores in the pad 108 are full of the gel-like material to provide the electrically conductive path from the shoe 104 to the contaminated surface 120. The insulating handle 112 and stand-off 116 are typically formed from the same insulating material, which is commonly rubber or plastic. The apparatus 100 may include one or more activators or switches to activate the pump 220 to pump gel-like material to the apparatus 100 and apply a voltage to the shoe 104. These functions may be activated by a common activator or separate activators.

[0046] The gel-like material supply line 204 is in communication with the pump 220 and decontaminating apparatus 100 to transport gel-like material under pressure from the reservoir 216 to the apparatus 100. The line 204 is preferable formed from a flexible tubing material to permit the user to move the apparatus 100 freely across the surface 220.

[0047] The gel-like material reservoir 216 and pump 220 are sized to accommodate the desired application. The pump typically has sufficient size to supply the gel-like material to the apparatus 100 at the desired rate, which is typically up to about 0.1 gal/min. The preferred pump types are peristaltic and diaphragm. An overpressure hydraulic loop (not shown), such as a recirculating hydraulic overpressure bypass across the pump 220 and a valve having a high cracking pressure, may be provided to prevent over pressurization of the supply line 204.

[0048] The first and second conductor cables 204 and 212 and power supply 224 are configured to cause the surface to be the cathode or the anode (as shown), depending on the composition of the surface 120 and the type of contaminants to be removed. As well it should be appreciated, cathodic stripping occurs when the surface is configured as the cathode, and anodic stripping when the surface is configured as the anode. In cathodic stripping, reduction occurs at the surface, and, in anodic stripping, oxidation occurs at the surface. Thus, in cathodic stripping metal oxides are stripped from the surface, and, in anodic stripping, metals are stripped from the surface. The power supply may be AC or DC and typically has voltage ranging from about + or -1 to about + or -15 Volts.

[0049] FIG. 3 is a hardware schematic further depicting the system 200. In addition to the components discussed above, the system 200 includes a processor 300, a memory 304, a power control module 308, and a current sensor 312. The processor 300 and power control module 308 collectively effect the temporal application of a desired voltage waveform to the first and second conductors 204 and 212. The processor 300 uses the sensed voltage (between the shoe 104 and surface 120) from voltage sensing lines 312 and 316 and the sensed current from the current sensor 320 (which is shown as a shunt resistor) from signal lines 324 and 328 to account for drift in the applied voltage waveform and

thereby maintain optimal operating conditions. As will be appreciated, the processor **300** may also control other operations, such as system alarms and safety features for abnormal conditions (e.g., current flow or short circuit) and a system display (not shown) and receive user commands via user interface devices, such as a keypad and switches (not shown). The processor **300** may look up a library of optimized waveform programs stored in the memory **304** for differing surface materials/contaminants. For example, a stainless steel contaminated surface would have a first optimal voltage waveform, a carbon steel surface a second (different) optimal waveform, and an aluminum surface a third (different) optimal waveform. The processor may control the pump **220** to ensure gel-like material introduction to the shoe **104** at an optimal rate preventing excess waste generation. The processor can display useful operating information, such as elapsed time, applied current and/or voltage, and quantity of gel-like material dispensed.

[0050] The time-dependent voltage waveforms used during electrolysis can have a profound impact on the stripping and contaminate removal rates. **FIGS. 8 and 9** show two possible voltage waveforms. When the constant voltage waveform **900** of **FIG. 9** was applied, the current waveform **904** would quickly ramp upwards to a peak **908** and then drop to a lower threshold value. The most effective surface removal commonly occurs during the ramp-up and short period of elevated current. While not wishing to be bound by any theory, factors believed to contribute to the quick decay of the current include polarization of the gel layer, water molecules in the gel-like material forming an organized dipole layer on the contaminated surface, and electropolymerization occurring at the anode. In contrast, when a cyclic waveform, such as the waveform **800** of **FIG. 8**, was applied, the current waveform **804** would ramp up **808** to an elevated level and substantially maintain the elevated level in later voltage application cycles. Effective surface removal occurred through the use of a cyclic voltage waveform. Although a square-wave voltage waveform was used, it is to be understood that a symmetric or asymmetric cyclic waveform of any shape maybe employed. The preferred frequency of the waveform ranges from about 5 to about 30 Hz. The applied voltage preferably ranges from about + or -2 to about + or -2.5 V. The voltage maybe Direct Current, Alternating Current, or a combination thereof.

The Gel-Like Material Composition

[0051] The gel-like material has a number of desired properties. For example, the material preferably is at least moderately conductive to provide sustained electrochemistry, generates no or only a small amount of non-hazardous off-gas, has a viscosity that is high enough to have no more than a specified amount of sloughing during application to the surface **120** but low enough to be pumped through the supply line **204** at the required rate, can cure in both molecular oxygen-containing and inert atmospheres, has a cure time ranging from about 1-8 hours, when cured has sufficient strength, elasticity, and tear resistance to be removed by hand in its entirety with little, if any, residue remaining on the decontaminated surface **120**, has a high (more preferably at least about 90%) rate of removal not only of smearable but also of fixed contamination from ferrous and nonferrous metals such as iron and stainless steel and aluminum, copper, tin, nickel, lead, titanium, zirconium, and hafnium and alloys thereof, does not generate hazardous

waste as determined by the TCLP for EPA RCRA metals (method SW-846), encapsulates contaminants and thereby mitigates airborne contamination, retains removed contaminants throughout the decontamination process, and, in the uncured state, the gel-like material has a purplish or magenta color and in the cured state, the material has a mustard yellow color (in U.S. nuclear color programs magenta purple color typically refers to wet radioactive-contaminated paint while mustard yellow color denotes radioactively contaminated plastic). More preferably, the uncured gel-like material has a viscosity ranging from about 1,000 to about 100,000 cPs, and sufficient conductivity to provide current densities at the contaminated surface of from about 0.1 to about 5 amps/inch² and the cured gel-like material has a tensile strength ranging from about 200 to about 400 psi.

[0052] To provide these properties, the gel-like material includes a number components, namely a latex formulation, an electrolytic agent, a thixotropic and/or viscosity enhancing agent, and a chelating or complexing agent. The material typically includes from about 83 to about 99 wt. % of the latex formulation, from about 1 to about 12 wt. % collectively of the electrolytic agent and thixotropic and/or viscosity enhancing agent, and from about 0 to about 5 wt. % of the chelating or complexing agent.

[0053] The latex formulation provides the desired strength, flexibility, and elastic properties of the cured gel-like material. The latex formulation is commonly an emulsion of insoluble monomers and surfactant in water. The typical latex formulation includes from about 20 to about 60 wt. % monomer, from about 2 to about 10 wt. % surfactant, and from about 0.1 to about 1.0 wt. % initiator, with the balance being water. When the initiator decomposes in the presence of radiation, it generates reactive species which, in turn, react with the monomers in the latex formulation (or with the separate monomer phase (micelles)), to initiate the desired polymerization. As will be appreciated, the cured latex can include any number of natural or synthetic oligomer, polymer, and copolymers derived from monomers, such as rubbers, esters, nylons, acrylates, acetates, butadienes, acrylonitriles, amides, carbonates, ethylenes, acrylics, vinyls, and the like. The uncured latex formulation typically contains monomers of one or more of these polymers. In the presence of an initiator, the latex formulation polymerizes to provide the oligomers and long-chain polymers. The long-chain polymers form a backbone imparting strength, flexibility, and elasticity to the cured gel-like material. Preferably, the latex formulation is a mixture of monomer types. More preferably, the latex formulation comprises from about 50 to about 75 wt. % KATS 9050TM (a proprietary vinyl formulation manufactured by the Whitmore GroupTM and from about 10 to about 50 wt. % VCI-372TM (a proprietary acrylic formulation manufactured by CORTEC, Inc.TM).

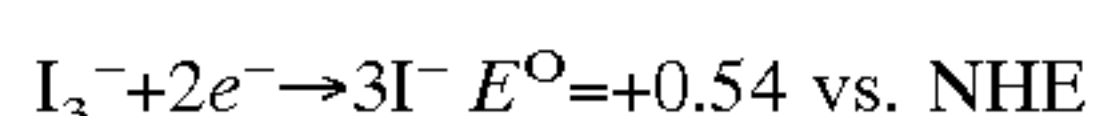
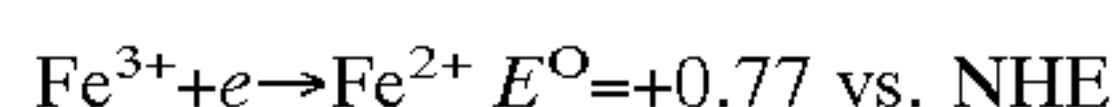
[0054] Preferably, an oxidation/reduction potential (ORP) of the electrolytic agent that is greater than that of the contaminant and/or decontaminated surface component material that is to be stripped but less than the ORP above which the other organic components of the gel-like material are oxidized by the electrolytic agent. In one application for removing plutonium from stainless steel surfaces, the ORP of the agent ranges from about 0.50 to less than 1.70. This upper ORP Bound (1.7 volts) is the ORP for potassium permanganate which is known to oxidize latex components

in the gel-like material. Typically, the agent is a metal or metal compound, with the metal preferably being selected from Groups 6-12 and 17 of the Periodic Table of the Elements and more preferably being a halogen or transition metal, with iron and iodine-containing compounds being preferred and iodine and iodine-containing compounds being most preferred. As will be appreciated, both iron and iodine species are nontoxic. As will be further appreciated, other acids, bases or salts can be added to the material to enhance further conductivity. For example, halides (e.g., chloride, bromide, and fluoride) and nitrate and sulfate ions may be used as the electrolytic agent but the halides can oxidize to form hazardous gases, such as chlorine, bromine, and fluorine), and nitrate and sulfate ions can reduce to form gaseous oxides of nitrogen and sulfur, respectively, such as NO_x and SO_x .

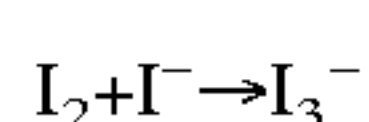
[0055] The thixotropic and/or viscosity enhancing or thickening agent preferably is present in an amount sufficient to provide the viscosity and shear rate noted above. Typical thickening agents include gums, alkyl esters of methacrylic acid, polyisobutylenes, clays such as bentonite, starch, and mixtures thereof. In one formulation, the electrolytic and thickening agents are the same compound. In this formulation, the compound is iodine and/or an iodine-containing compound. The particularly preferred agent is a mixture of MI , I_2 , and I_3^- , where M is a metal, such as sodium and potassium. In one configuration, the agent comprises from about 0.5 to about 1.0 mole % MI and from about 0.25 to about 0.5 mole % I_2 , with the remainder being water.

[0056] The chelating or complexing agent reacts with metal cations in the gel-like material and inhibits the metal cations solubilized in the gel-like material during stripping from being removed from the gel-like material. The metal cations are typically the contaminant metals stripped from the contaminated surface. The complexing agent is not believed to react with the preferred electrolytic and thickening agent(s) because the agents either have no charge or are anions. Although any complexing agent may be employed, the preferred complexing agent is EDTA.

[0057] In applications where hydrogen gas is generated at the cathode a molecular hydrogen suppressant may be included. The suppressant reduces preferentially relative to water and thereby prevents the generation of molecular hydrogen. The suppressant is preferably a halogen or a transition metal, with iron and iodine-containing compounds being preferred. Iron (m) reduces to iron (II) and iodine reduces to iodide in accordance with the following reactions:



[0058] Iodine is stabilized in water as a complex with iodide ion by the reaction:



[0059] In applications where oxygen gas is generated at the cathode a molecular oxygen suppressant may be included. The suppressant oxidizes preferentially relative to water and thereby prevents the generation of molecular oxygen. The preferred molecular oxygen suppressant is oxalic acid and/or oxalate (depending on the pH). The oxidation of the suppressant occurs according to the following reaction:



[0060] Carbon dioxide is a nontoxic gas and poses no hazard to equipment or personnel working in ventilated areas because the quantity of carbon dioxide gas produced is typically small (e.g., 4.58 ml/min CO_2).

[0061] The amount of the molecular hydrogen and oxygen suppressants is preferably at least the stoichiometric amount required to inhibit molecular hydrogen or oxygen generation, respectively, and more preferably ranges from about 0 to about 0.03 wt. % of the gel-like material.

Manufacture of the Gel-Like Material

[0062] The manufacture of the gel-like material will now be described with reference to FIG. 7.

[0063] In step 700, the selected latex formulations are mixed in a stirred vessel to form a latex mixture 704.

[0064] In step 708, an aqueous iodine-containing solution is mixed in a separate stirred vessel with iodide crystals to form an iodine mixture 712. They are stirred for a sufficient period of time for the crystals to be dissolved completely in the aqueous iodine-containing solution. During mixing a portion of the iodine reacts with the iodide to form tri-iodide ion according to the equation referenced above.

[0065] In step 716, the iodine and latex mixtures 712 and 704 are combined in a stirred vessel. The rate of addition of the iodine mixture 712 to the latex mixture 704 can be important. If the iodine mixture 712 is added too rapidly, the iodine-containing components in the iodine mixture 712 can react with and destroy the monomers in the latex formulation(s). By adding the iodine mixture 712 slowly in the presence of agitation, it is believed that the mixture 712 is dispersed as small granules or droplets in the latex mixture 704. The rate of addition of the iodine mixture 712 to the latex mixture 704 must not be sufficient or high enough to oxidize the latex mixture 704.

[0066] In step 720, the chelating agent is added to the combined mixture. Step 720 may be performed before, during, or after step 716. When performed before step 716, the chelating agent is added to the latex mixture 704.

[0067] In step 724, the various components are stirred to form the gel-like material 728. Mixing is complete when the color of the gel-like material is magenta rather than white.

The Decontamination Process

[0068] The decontamination process will be described with reference to FIGS. 4 and 5A-E.

[0069] In step 400, the porous and permeable abrasive pad 108 is engaged with the scrubbing shoe 104.

[0070] In step 404, the gel-like material 124 is applied to the contaminated surface 120 by being pumped to the decontaminating apparatus 100 and through the pad 108 onto the surface 120. The step is illustrated by FIG. 5A. The gel-like material 124 is spread as a relatively smooth layer onto the surface 120. As the material 124 is applied to the surface 120, the decontaminating apparatus is moved in circular motions parallel to the plane of the surface 120 to mechanically abrade and apply the gel-like material 124 uniformly to the surface.

[0071] In step 408, when the gel-like material layer 124 has the desired thickness on the surface 120, as shown in

FIG. 5B voltage is applied by the power source **224** to the decontaminating apparatus **100**, causing electric current to flow from the shoe **104**, through the gel-like material **124**, and to the surface **120**. During the application of voltage, the apparatus **100** is continuously moved in the circular motions around the surface **120** in the same manner noted above. The circular motions help establish improved electrical contact through minor abrasion, help break through surface deposits, such as oil and grease, and stir the gel-like material to diffuse contaminants already entrained in the gel-like material **124**. During this step, it is desirable that the gel-like material layer **124** not be too thick to interfere with the flow of current and/or proximity of the shoe **104** to the surface **120**. The desired thickness of the gel-like **124** layer in this step ranges from about 0.015 inches to about 0.025 inch. As will be appreciated, stainless steel contains about 18 wt. % chromium and 11 wt. % nickel, with the balance being iron. Typically where stainless steel is the surface **120**, in step **408** iron is primarily stripped along with lesser amounts of chromium and nickel. Surface contaminants, such as plutonium, are transferred to the gel-like material **124** through mechanical abrasion and agitation, where it reacts with the chelating agent.

[0072] In step **412**, when the surface **120** is stripped by the desired amount additional gel-like material **124** is applied to the layer and spread uniformly across the surface using a trowel edge **500** of the shoe **104** as shown in **FIGS. 5C and 5D**. This provides a layer of sufficient thickness to permit efficient post-cure removal. The desired thickness of the gel-like layer **124** after this step ranges from about 0.025 to about 0.035 inch (cured).

[0073] In step **416**, the gel-like material is allowed to cure to form a cured gel-like material layer **504**. The gel-like material is typically fully cured in about 1 to about 8 hours and converts to a yellow film.

[0074] In step **420** and as shown by **FIG. 5E**, the cured gel-like material **504** is removed from the decontaminated and electro-polished surface when desired, leaving a clean and decontaminated surface exposed. Little, if any, of the cured gel-like material remains on the surface **120**. As shown in **FIG. 6**, the cured gel-like material **504** includes the removed contaminants **600** locked in the material **504**. The removal of the gel-like material layer is by peeling, which may be initiated with the trowel-edge **500** or a scraper edge or by using friction from a gloved finger.

Experimental

[0075] Tests were performed in an electrolysis cell to determine voltage-current behavior and off gas generation. Power was supplied to the cell at a constant voltage to induce a direct current that was then measured as a function of time for the various candidates. Tested electrolyte solutions are given in Table 1.

TABLE 1

Electrolyte Solutions Tested		
Test	Species	Condition
1	Sulfuric Acid	Adjusted to PH = 2
2	Oxalic Acid	Adjusted to PH = 2
3	Ferric Sulfate	1 M
4	Iodine/Iodide Complex	1 M

[0076] For each solution, the electrolytic cell head-space was purged with helium gas, and a voltage between 2 and 6 VDC was then applied across the anode and cathode. Electrical current and gas evolution from each electrode were noted. Table 2 shows a summary of the results.

TABLE 2

Summary of gas evolution experimental results.			
Electrolyte	Current	Cathode Gas	Anode Gas
Deionized water	11-71 μ a	—	—
Sulfuric acid (pH = 2)	10-825 ma	Hydrogen	Oxygen
Oxalic acid (pH = 2)	1-2000 ma	Hydrogen	Oxygen
			Carbon Dioxide
Ferric sulfate (1 M)	50-1350 ma	—	Oxygen
Iodine/Iodide (1 M)	30-320 ma	—	Oxygen

[0077] These tests showed that the presence of either ferric ion or iodine in the electrolyte eliminates the production of hydrogen at the cathode, as expected. Oxalic acid does not completely eliminate the production of oxygen at the anode but reduces the rate of production to less than 1.5 ml/min. Based upon these results, oxalic acid and iodine/iodide (along with other additives) can be used able to decontaminate electrolytically stainless steel surfaces without generating hydrogen or other hazardous gases. Carbon dioxide and oxygen are the only off gases produced-in very low quantities.

[0078] The iodine/iodide formulation was chosen for further electrochemical testing. To begin, tests were performed to find an optimum electrochemical activity point using various additives. This work showed that iodine needed to be present to strip surface contaminants from test coupons. Cyclic voltammetry data indicated that potassium iodide alone could accomplish the desired stripping, but, when tested, solutions containing only potassium iodide failed to produce the anticipated result. It is believed that reactions in the aqueous solutions form iodine that becomes active during the stripping process, and, in a gel-like material, these reactions occur much more slowly. A series of solutions were prepared with increasing concentrations of iodine. Table 3 shows the concentrations used, and the time it took to decontaminate the sample coupons.

TABLE 3

Test for Optimal KI/I2 Concentration For Electrochemistry.			
Solution	KI (ppm)	I2 (ppm)	Complete/Partial Stripping Time (sec)
1-5	Test	—	—
6	1350	675	25/12.5
7	2700	1350	20/11.1
8	5400	2700	12.5/10
9	10800	5400	12.5/10
10	21500	10800	10/not tested

[0079] Increasing the concentration of iodine in solution only slightly decreased the stripping time. It was determined that achieving the physical gel-like material characteristics required for ease of stripping while maintaining the proper thixotropic properties needed to prevent flowing of the material were more important than minor reductions in stripping time. Subsequent tests also showed that electrical phenomena had a more pronounced effect on cleaning time

than iodine concentration. Solution 10 is not viable because iodine begins to come out of solution leaving beads of iodine visible, and the gel-like material is nearly solidified fully at this concentration.

[0080] Solutions 6-10 were tested for drying time and their post-cure physical properties. Acceptable gel-like material formulations would not be brittle when cured nor excessively stretchy when peeled away from surfaces. Solutions 9 and 10, having high iodine concentrations, were very stretchy and required long times to cure. Solutions having lower iodine concentrations, such as solution 6, were excessively brittle. **FIG. 10** shows solutions with increasing KI/I₂ concentrations and their relative viscosities. Solution 8 uses a lower concentration of iodine and showed an acceptable ability to stay in place on vertical surfaces. In addition, it was less stretchy and not brittle post-cure. Considering these factors, it was chosen as the optimal gel-like material recipe and additional testing was initiated.

[0081] Because all elements in the actinide series are radioactive, none of them can be considered for use as a possible radioactive plutonium surrogate. However, elements in the lanthanide series are not radioactive but have the same electron configuration as those in the actinide series, therefore exhibiting similar chemical properties. Not surprisingly, elements in this series also have similar oxidation states to the actinides. In particular, cerium exhibits both +3 and +4 oxidation states, which are common oxidation states of plutonium. Iron III oxide also has similar chemistry to plutonium and other transuranium metals. Fe(III) is a hard acidic metal ion much like plutonium. When exposed to air, cerium forms cerium (III) oxide (Ce₂O₃) and cerium (IV) oxide (CeO₂) and iron forms iron (E) oxide (Fe₂O₃) and iron (II) oxide (FeO). Because of the similar chemical properties and lack of radioactivity, cerium and iron were chosen as safe and affordable plutonium surrogates for laboratory testing.

[0082] Laboratory test equipment was fabricated to test candidate electrolyte formulations and various additives. The test system included a DC power supply, electrochemical analyzer, voltage and current meters, and an electrochemical cell assembly. The cell assembly was comprised of a working electrode, i.e. the test coupon, a counter electrode, reference electrode, gas purge port, and condenser.

[0083] Working electrodes were fashioned either from 304 stainless steel rod attached with epoxy adhesive to a glass cylinder, or 304 stainless steel sheet 2 mm thick. A platinum coil was used as a counter electrode, while an Ag/AgCl electrode served as a reference electrode. For comparison with published electrochemistry data in the literature, a second working electrode was made from platinum rods sealed in Teflon. All electrodes were fabricated from 16-mm diameter rod.

[0084] In initial experiments, copper and silver were very effectively removed from stainless steel plates, demonstrating that the fundamental concept of electrodecontamination was feasible. The electrochemical behavior of these plated metals is well known, and this fact permitted their detailed study for comparison to the literature. Using a simple electroplating process, copper was reduced onto stainless steel plates from solutions of copper sulfate. By designing the process of the present invention to completely remove "contaminants" chemically plated onto a steel substrate, it was hypothesized that less-well bound contaminants would be readily removed. This hypothesis proved correct in laboratory tests when cerium, deposited onto stainless steel

plates as a surrogate contaminant, was successfully removed. To prepare the test coupons, a paste of cerium metal powder and mineral oil was smeared onto the stainless steel (304) coupons and baked at 500° C. for seven hours.

[0085] The experimental set-up used to test the effectiveness of the process of the present invention included an aluminum cathode "terminal," with the anode comprised of a stainless steel plate. Nylon (non-conducting) screws held the plates together, sandwiching a silicone gasket in between. This space between the anode and cathode faces was held constant, effectively controlling the gel-like material layer thickness. For testing, the "contaminated" metal plate (anode) was electrically connected to the positive terminal of a DC power supply, while the aluminum cathode was connected to the negative side. Testing was done with an electrochemical analyzer designed to work with three electrodes. As the system of the present invention has only two electrodes, the terminals for the analyzer reference and counter electrodes were shorted together and connected to the aluminum plate, which then served as the counter electrode. The working electrode was the stainless steel plate. A quantity of gel-like material was applied to the treated metal plate, and the cathode was then placed on top to form a "sandwich" to pass electrical current. Initially, a constant voltage between 2 to 12 VDC was applied to the sandwich for a period of time ranging from 1 to 10 minutes. At the conclusion of the test, the aluminum block was lifted away and the patch of expended electrolyte on the anode was allowed to cure, usually requiring 1 to 3 hours. Following complete curing of the gel-like material, the patch was peeled away to determine how well the surface had been cleaned.

[0086] The decontamination process was improved by introducing a ScotchBrite™ pad Type 7446™ between the anode and cathode. The pad consists of polymer strands impregnated with an inert abrasive specially designed to break up oxidized material on stainless steel substrates. Each pad was saturated with electrolyte gel-like material prior to testing. A gentle circular motion was used during some of the tests to loosen oxidation scale on the metal surface. Testing showed that the thickness of this pad created a wider gap (~1 cm) than was optimal for the process of the present invention. With the ScotchBrite™ pad in place, the initial current was 60 mA, and without the pad it was 80 mA. Tests were run with various materials having less thickness, but, as shown in Table 4 below, these significantly decreased the current as well. Finally, it was discovered that 3M™ manufactures a thin version of the ScotchBrite 7446™ pad, only ~4 mm thick, which did not adversely affect the current. A continuous current density of 1 to 1-½ amps per square inch, applied for at least 30 seconds, was found to adequately remove surface contamination, with an applied unregulated voltage of 12 VDC. This current density is less than had been expected, with the reduction believed to be attributable to the excellent electrolyte conductivity and agitation of the gel-like material during the process.

TABLE 4

Test for scrubber material resistance to current.	
Material	Current (A)
No Mesh	1.12
Polypropylene Square Mesh, 0.03" × 0.037" Opening	0.63
Polypropylene Square Mesh, 0.120" × 0.175" Opening	0.32
Polyethylene Diamond Mesh Nominal 0.15" Opening	0.51

TABLE 4-continued

Test for scrubber material resistance to current.	
Material	Current (A)
Monofilament Nylon Mesh, 9.1 × 9.1 Mesh, 0.0787" Opening	0.71
Monofilament Polyester Mesh, 7.9 × 7.9 Mesh, 0.0937" Opening	0.54
Monofilament Polypropylene Mesh, 0.0661" Opening	0.65

[0087] Electrochemical tests on the gel-like material showed that the iodine/iodide component actively strips the contaminated surface, while remaining components cause entrainment of the contamination and curing of the gel-like material. Tests to find the optimal concentration for iodine were performed, and the results showed that decontamination was generally not a function of the concentration of iodine/iodide—but if no iodine/iodide was present then the electrochemical decontamination was not successful. Tests also showed that varying the concentration affected the viscosity of the gel-like material, so that a high concentration created a solidified gel-like material, while a low concentration created a low-viscosity gel-like material.

[0088] It was observed during testing that the DC current would quickly ramp upwards to a peak, 1-½ amps/inch², and then drop to a lower threshold value, 300 mA/inch², where it would remain. In addition, it appeared that the most effective surface removal during the process occurred during the ramp-up and short period of elevated current. If the applied voltage was removed and quickly reapplied, the current would again peak and decline as before.

[0089] The rapid initial material removal and subsequent current decline strongly suggested that a time-dependent phenomenon was occurring and that a cyclic applied voltage could potentially be used to maximize the process effectiveness and efficiency. Initial testing to determine if this would be effective was done using a square-wave voltage, which was applied to the electrolytic cell with varying duty cycles and asymmetric values centered about zero (e.g. +6 VDC for ½ time switching to -2 VDC for 2/3 time) in an attempt to discover if any combination would yield a marked performance improvement. A summary of the results is shown in Table 5.

TABLE 5

Applied cyclic power waveforms and test results.	
Waveform	Result
+12 VDC, -12 VDC symmetric square-wave applied at 10 Hz	No cleaning effect. Final electrolyte appeared "scorched" and would not readily peel when cured.
+12 VDC, 0 VDC sine-wave (full-bridge rectification) at 60 Hz (line power).	Rapid cleaning, ill-defined edges on cleaned area, no adverse effects on gel-like material cure (this combination was used in the prototypes).
+12 VDC ½ time, -3 VDC ½ time symmetric square-wave applied at 10 Hz.	Cleaning action occurred but noticeably slower. Sharp definition of cleaned areas. No adverse effects on gel-like material cure.
12 VDC for ⅔ time, -3 VDC for ⅓ time.	Very sharp edges delineating cleaned patch. Slightly slower cleaning rate. No adverse effects on gel-like material cure.

TABLE 5-continued

Applied cyclic power waveforms and test results.	
Waveform	Result
+6 VDC for ¾ time, -1.5 VDC for ¼ time.	Odd, sharply defined clean "streaks" under gel-like material patch, adjacent regions unaffected. No adverse effects on gel-like material cure.

[0090] After determining that the electrochemical behavior of the electrolyte gel-like material could be affected by varying the imposed power waveform, a more comprehensive study into the behavior of the electrolyte components was initiated. Using the test cell described above, the gel-like material constituents were tested in aqueous solutions to determine their electrochemical activity.

[0091] Cyclic voltammetry is a process used by electrochemists to verify that a reaction will occur at certain voltages. This process was used to verify that the gel-like material constituents could oxidize metals on the surface of a stainless steel plate.

[0092] All of the voltammograms with iodide or iodide-iodine mixtures present show two distinct peaks for iodide oxidation and iodine reduction on the platinum and stainless steel electrodes. These tests showed an iodine reduction peak at around -0.3 V vs. Ag/AgCl and an iodide oxidation peak at around 1.55 V vs. Ag/AgCl, see FIG. 11. Copper stripping and deposition peaks are also present on platinum and stainless steel, and the copper stripping voltage (-0.25 V vs. Ag/AgCl) coincides with the iodine reduction peak on the stainless steel electrode, suggesting that iodide oxidation via production of iodine may also be involved in the copper stripping process.

[0093] Tests on the gel-like material with and without iodine showed that iodine should be initially present for stripping to occur. The peaks for copper stripping were verified by tests with a solution of CuSO₄ and H₂SO₄, which showed a peak around -0.3 V vs. Ag/AgCl.

[0094] Tests were also run with various components forming the gel-like material matrix, including acrylonitrile and vinyl acetate. Other components were tested, but no significant information was gained from the analyses. Cyclic voltammograms of all of the polymer gel-like material constituents used in the formulation of the gel-like material electrolyte were recorded in aqueous 0.1 M NaCl. FIG. 12 and FIG. 13 illustrate that acrylonitrile and vinyl acetate are electrochemically active.

[0095] When cyclic voltammetry was recorded in the presence of KI/I₂ mixture, the current was two orders of magnitude higher (10⁻² A) and the peaks depicted only oxidation of iodide and reduction of iodine as in FIG. 11.

[0096] The current-voltage curve recorded in the test cell with gel-like material electrolyte and a coated copper coupon is shown in FIG. 14. Anodic and cathodic waves are seen, which are most likely due to iodide oxidation and iodine reduction. The iodine oxidation peak at 2.2 V vs. Ag/AgCl was used for initial testing of voltage waveforms.

[0097] Cyclic voltammograms recorded in aqueous KI/I₂ solutions, show that copper stripping and deposition peaks were much smaller than the iodine reduction and iodide oxidation peaks. Therefore, it is concluded that a wave in

that range 1.8-2.4 V will cover iodine oxidation and copper stripping. The stripping experiments show that copper stripping occurs in the voltage range 2.1-2.4V.

[0098] FIG. 15 illustrates the voltammogram obtained in the test cell, with iron oxide as the working electrode contaminant. It can be clearly seen that the wave for iodide oxidation is at 1.4 V and the wave for iodine reduction is between -1.6 and -3.0 V. It is also seen that there is a slight oxygen evolution at 3 V, and no hydrogen evolution. Reduction is needed to remove the oxide from the surface, so initial voltage waveform testing was conducted between -1.6 to -3.0 V.

[0099] Chronoamperometry is an electrolysis process performed at a constant voltage potential. In one-step chronoamperometry, the process slowly steps down the current at a constant rate so that current-time transients in the process can be viewed.

[0100] To test the voltage and find the time necessary for copper stripping in the cell, a DC voltage was applied and the current-time transients monitored. Copper was deposited onto 2-cm×3-cm coupons in a 100-ml cell with two stainless steel electrodes 1.5 cm apart. Copper deposition was performed in 0.05 M CuSO₄/0.1 M sulfuric acid at -2.2 V. Copper coated stainless steel coupons served as the working electrode, and an aluminum plate was used as the counter (negative) electrode. Under these conditions, the applied voltage was initiated at 1.8V and increased to 2.4V over time. A typical current-time curve, the response to a constant applied voltage, is seen in FIG. 16. The transient observed is due to copper being stripped from the anode.

[0101] After the reaction was finished, polymerized gel-like material was attached to the anode, indicating the transient curve actually covers copper electrostripping and anodic gel-like material polymerization. Results were best at a stripping voltage of 2.4 V at 300 to 600 seconds, but further testing was needed to eliminate the gel-like material polymerization.

[0102] The same process was used to evaluate iron oxide stripping. The electroreduction of iron oxide proceeded best at -3.0 V; a typical transient curve for the electroreduction of iron oxide is seen in FIG. 17. No gel-like material polymerization was seen in the iron oxide stripping tests, so the transient is due only to iron oxide electrostripping reactions. These time transients should give an indication of the time needed to strip the contaminated surface, and so initial testing on metal oxide stripping will be performed for less than 100 seconds as indicated in FIG. 17.

[0103] Square wave voltammetry uses a constant-voltage waveform, and is most often used for analytical purposes; in this case it was used to enhance the electrostripping process. A representative waveform is depicted for the oxide stripping tests in FIG. 18. The waveform can be modified, increasing or decreasing its voltage, as well as the voltage range, increment size, and amplitude.

[0104] Voltammetry tests with copper "contaminant" used the parameters given in Table 6 for copper stripping.

TABLE 6

Parameters used in square wave voltammetry tests for copper.	
Initial Voltage (V)	2.3
Final Voltage (V)	2.1
Increment (mV/s)	1

TABLE 6-continued

Parameters used in square wave voltammetry tests for copper.	
Amplitude (mV)	2
Frequency (Hz)	10
Sensitivity (A)	0.1

[0105] It was assumed that ion transport in the system of the present invention with gel-like material as the electrolyte would be slower than in an aqueous system, so a slow voltage increment and amplitude change were chosen. The voltage range was based on the previous test and the cyclic voltammetry data. A voltage range of 1.8-2.0 V showed almost complete stripping.

[0106] The test was repeated multiple times, and in all the experiments copper was completely stripped off in 20 seconds or less at 10 Hz. Parametric measurements were made to find the best conditions for copper electrostripping using square wave voltammetry, see Table 6 and Table 7.

TABLE 7

Metal stripping vs. voltage range using square wave voltammetry.	
Voltage Range (V)	Stripping Effect
1.7-1.9	None
1.8-2.0	Stripped
1.9-2.0	None
2.0-2.2	Partial
2.1-2.3	Partial
2.2-2.4	Partial

[0107] After these tests were complete, the voltage range was set from 1.8 to 2.0 V with all other parameters the same as those given in Table 6. The frequency was varied in these tests, which affected the time required for stripping. As the time decreased, the stripping of copper also decreased.

TABLE 8

Stripping efficiency vs. time and frequency.		
Frequency (Hz)	Time (sec)	Stripped
10	60	Yes
12.5	48	Yes
15	40	Yes
20	30	Partial
25	24	Partial

[0108] In Table 8, the higher frequency (20-25 Hz) tests needed more than 30 seconds to strip the copper films, whereas the lower frequency of 10 Hz was able to strip in 20 seconds. It was determined that a metal contaminant could be stripped with oxidizing potential voltage of 1.8-2.0V and waveform oscillation period of 0.1 sec, i.e., a frequency of 10 Hz. Oscillation between voltages decreased the time needed to strip by a factor of ten. Previous tests required 10 minutes or more to remove copper films at a set voltage of 2.4V.

[0109] Iron oxide was stripped over a wide voltage range at a relatively low frequency in 200 seconds; the parameters are shown in Table 9. The waveform for this analysis

resembled that in **FIG. 18**. All of the initial tests in the range -2.7 to -3.5 V showed complete stripping of the iron oxide thin film, so the highest voltage window was chosen to find the shortest time needed to completely strip iron oxide as shown in Table 10.

TABLE 9

Parameters used in square wave voltammetry tests for iron oxidestripping.	
Initial Voltage (V)	-3.0
Final Voltage (V)	-3.5
Increment (mV/s)	1
Amplitude (mV)	2
Frequency (Hz)	2.5
Quiet time (sec.)	20
Sensitivity (A)	0.1

[0110]

TABLE 10

Time and frequency for iron oxide stripping.		
Frequency (Hz)	Time (sec)	Stripped
5	100	Yes
7.5	66	Yes
10	50	Yes
12.5	40	Yes
15	33	Yes
17.5	29	es
20	25	Yes
22.5	22	Partial

[0111] Electroreduction in the frequency range of 5-20 Hz resulted in complete electrostripping of the oxide layer. Partial electrostripping at 22.5 Hz indicated that 22 seconds was not sufficient under the given conditions for the process to completely decontaminate the surface. Therefore, it was determined that the lowest reduction time needed to strip iron oxide contamination occurred at a frequency of 20 Hz, or period of 0.05 seconds.

[0112] Minimum stripping times were not determined for all of the frequencies, but at 20 Hz a stripping time of less than 10 seconds was achieved, see Table 11. The minimum time needed to strip at 22.5 Hz was greater than 20 seconds, proving that lower frequencies strip more quickly. It was again shown that an oscillating voltage decreased the time need to strip about ten-fold, going from minutes to seconds. It is clear that square wave voltammetry accelerates the electrostripping process within the gel-like material.

TABLE 11

Minimum stripping time needed for 20 Hz frequency square wave tests.		
Increment mV	Time (sec)	Stripped
2	12.5	Yes
30	8.33	Yes
40	6.25	Partial

[0113] It is worth noting that it was expected that the iron oxide would be reduced onto the metal surface, and not become entrained in the gel-like material, but in a test after the electrochemical process was performed the gel-like

material could be rinsed away leaving behind the iron contaminated surface. Afterwards the iron oxide could be rubbed off of the surface, which was not possible before the electrochemical process. Also, when this reducing process was utilized the two plates showed 80 and 92% removal of the fixed waste.

[0114] Chronoamperometry of iron oxide, run from -3 to -3.5 V using 300 steps and a frequency of 20 Hz resulted in complete reduction of the iron oxide film (a surrogate for plutonium oxide) in just 15 seconds.

[0115] The parameters and waveform are shown in **FIG. 19** and Table 12 below. This waveform will also work over the voltage range 1.8 to 2.0 V to strip contaminants such as copper or plutonium.

TABLE 12

Parameters for chronoamperometry.	
Initial Voltage (V)	-3.0
High Voltage (V)	-3.0
Low Voltage (v)	-3.5
Number of Steps	300
Pulse duration (sec)	0.05
Sampling rate (sec)	0.1

[0116] This experiment showed that multiple-step chronoamperometry is an excellent method for enhancing the electrochemical stripping process. This method is easier to apply continuously, such as in the electrodecontamination of surfaces.

[0117] Five criticality barriers previously used in fuel storage basins at nuclear test facility were used for this experiment. At each spent nuclear fuel storage port in the large fuel storage racks, a criticality barrier is used as a cover of the fuel rod storage location and aids in preventing putting fuel into a port that is already loaded. This is not the primary line of defense against overbatching fuel ports (numerous checks and verifications are employed), but because of its function it is termed a criticality barrier. It is constructed of stainless steel plate and has dimensions of approximately $14" \times 14" \times \frac{3}{8}"$. A hand held Geiger counter (Ludlum measurements) with a 2" diameter probe head was used to measure the surface radioactivity, before and after decontamination. The probe/heads of both analyzer were positioned approximately $\frac{1}{4}"$ away from the flat surface during radioactivity measurement. Filter paper swipe samples were also collected to provide an estimate of the level of loose contaminants on the test articles, before and after being decontaminated.

[0118] Between 5 and 10 minute cleaning applications were used to apply the electrolyte gel-like material, moderately scrub the surface, and pass current through the interface. A minimum of 2 hours of cure time was adopted prior to stripping the coating away from the surface. One plate, **3A**, was tested in a vertical application. Table 13 lists the test conditions, and the results of radioactivity survey on the surface of all five criticality barriers before and after decontamination. Based on swipe sample analyses, the major contaminants were ^{60}Co , ^{137}Cs and ^{152}Eu . However, the major contaminants were ^{60}Co , ^{95}Zr , ^{137}Cs , ^{152}Eu , ^{154}Eu and ^{155}Eu in the removed gel-like material samples.

TABLE 13

		<u>Evaluation Summary</u>				
		<u>Test Articles and Applied Current Parameters</u>				
Radioactivity Measurements (1)		2A 10 minutes, normal	3A 5 minutes, normal (3)	4A 5 minutes, reverse	5A 5 minutes, normal	6B 7 minutes, reverse
		<u>Pretest</u>				
Direct Scan Swipe Sample (dpm/100 cm ² (2)	Geiger Counter (βγ, dpm/100 cm ²) βγ	140,000 13,461	15,000 1,595	25,000 582	30,000 487	120,000 6,287
		<u>After Decontamination</u>				
Direct Scan Swipe Sample (dpm/100 cm ²) (2)	Geiger Counter (βγ, dpm/100 cm ²) βγ	5,000 37	5,000 22	5,000 3	3,000 29	10,000 51
Total % Removal	βγ (direct scan) βγ (swipe sample	96 99	67 99	80 99	90 94	92 99

Note:

(1) Background in hood: 50 μ R/hr, 2,000 dpm/100 cm² $\beta\gamma$ (2) α analyses of all swipe samples: <20 dpm/100 cm²

(3) Test article position at 60° angles from the horizontal surface

[0119] A similar test was conducted in 2002 to determine the performance of an earlier version of the system of the present invention. During those tests, an earlier version of the instrument removed an average of 86% of the fixed contaminants using 10 minute process time. While both units removed virtually all of the loose contamination, the improved version seems to remove more fixed contamination. Windex window cleaner and Bartlett's TLC Stripcoat (strippable coating) were also used as comparison methods (in the previous tests). These results are comparable to the current test because the same type of criticality barriers were employed. Table 14 lists the radioactivity measurements of the test articles, before and after decontamination with Windex and Stripcoat methods. Bartlett TLC Stripcoat is more effective than Windex cleaner to remove radioactive contaminants from the criticality barrier, but neither was as effective as the present invention.

[0120] Table 14. Radioactivity Other Decontamination Methods

TABLE 14			
Radioactivity Other Decontamination Methods			
Radioactivity Measurements		Test Article 1 (Windex)	Test Article 2 (Bartlett Strip Coating)
		Pretest	
Direct Scan	Geiger Counter ($\beta\gamma$, dpm/100 cm ²)*	16,000	20,000
	Surface Dosage (mR)	0.5	0.5

TABLE 14-continued

Radioactivity Other Decontamination Methods			
Radioactivity Measurements		Test Article 1 (Windex)	Test Article 2 (Bartlett Strip Coating)
Swipe Sample (dpm/100 cm ²)	$\beta\gamma^{**}$	1,160, 1,200	<1,000
	α^{**}	39, <20	<20
		After Decontamination	
Direct Scan	Geiger Counter ($\beta\gamma$, dpm/100 cm ²)*	13,000	9,000
	Surface Dosage (mR)	NA	NA
Swipe Sample (dpm/100 cm ²)	$\beta\gamma$	<1,000	1,000
	α	<20	<20
% Removal	$\beta\gamma$ (dpm/100 cm ² , direct scan)	19	55

*peak reading of the test article surface, fume hood floor has background of 3000 dpm/100 cm

**samples from different locations of test article

[0121] Other strippable coating tests were also tested. Table 14 shows the results of several other tests of similar types of strippable coatings. ALARA 11 46™ (Imperial Coatings™) and TLC Stripcoat™ (Bartlette Nuclear™) are soft, latex coatings that are popular for the removal of loose contamination. Their use for fixed contamination is limited (and typically not reported). PENTEK 604™ (PENTEK Co.™) is a chemical that incorporates organic acids and chelants and also is self stripping. It is designed to release from the substrate in about 24 hours and flake off of the surface. The INSTA-COTE™ test was a two step process that used a glycerin fog coating to capture the contaminants and the INSTA-COTE™ (Los Alamos National Laboratory), durable, quick drying coating to produce a surface that can be walked on in a very short time.

TABLE 14

Similar Tests of Other Strippable Coatings			
Type of Coating	Type of Contamination	Average Loose Containment Removal	Average Fixed Contaminant Removal
ALARA 1146 ²	Savannah River Fuel Fabrication Facility	82%	N/A
ALARA 1146 ³	SIMCON I and II	80%	58%
TLC Stripcoat ³	SIMCON I and II	77%	61%
PENTEK 604 ³	SIMCON I and II	93%	66%
TLC Stripcoat ⁴	NFS, Irwin. TN	88%	N/A
INSTA-COTE (and Capture Fog) ⁴	NFS, Irwin. TN	94%	N/A

[0122] As can be seen from the above, the other decontamination techniques tested were not as effective as the present invention in removing both fixed and smearable contamination.

[0123] A number of variations and modifications of the invention can be used. It would be possible to provide for some features of the invention without providing others.

[0124] For example in one alternative embodiment, the decontamination process can be used for applications other than radioactive decontamination. The process may be used to remove rust from carbon steel, leaving behind a near-ideal surface for welding. Iodine and iodide possess anti-viral and anti-bacterial properties. The gel-like material may therefore be used to remove decontamination from medical instruments, surgical masking, and surgical suites.

[0125] In another embodiment, the gel-like material includes one or more fire retardant additives. The additives inhibit or prevent the combustibility of the cured gel. The fire retardant additives are preferably intumescent fire retardants, such as phosphate ester. As will be appreciated, any number of other fire retardant additives may be used.

[0126] The present invention, in various embodiments, includes components, methods, processes, systems and/or apparatus substantially as depicted and described herein, including various embodiments, subcombinations, and subsets thereof. Those of skill in the art will understand how to make and use the present invention after understanding the present disclosure. The present invention, in various embodiments, includes providing devices and processes in the absence of items not depicted and/or described herein or in various embodiments hereof, including in the absence of such items as may have been used in previous devices or processes, e.g. for improving performance, achieving ease and/or reducing cost of implementation.

[0127] The foregoing discussion of the invention has been presented for purposes of illustration and description. The foregoing is not intended to limit the invention to the form or forms disclosed herein. In the foregoing Detailed Description for example, various features of the invention are grouped together in one or more embodiments for the purpose of streamlining the disclosure. This method of disclosure is not to be interpreted as reflecting an intention that the claimed invention requires more features than are expressly recited in each claim. Rather, as the following claims reflect, inventive aspects lie in less than all features

of a single foregoing disclosed embodiment. Thus, the following claims are hereby incorporated into this Detailed Description, with each claim standing on its own as a separate preferred embodiment of the invention.

[0128] Moreover though the description of the invention has included description of one or more embodiments and certain variations and modifications, other variations and modifications are within the scope of the invention, e.g. as may be within the skill and knowledge of those in the art, after understanding the present disclosure. It is intended to obtain rights which include alternative embodiments to the extent permitted, including alternate, interchangeable and/or equivalent structures, functions, ranges or steps to those claimed, whether or not such alternate, interchangeable and/or equivalent structures, functions, ranges or steps are disclosed herein, and without intending to publicly dedicate any patentable subject matter.

What is claimed is:

1. A method for treating an object, comprising:

(a) applying a gel-like material to a surface of the object;

(b) passing a current through the gel-like material on the surface of the object;

(c) curing the gel-like material to form a cured layer of the material; and

(d) removing the cured layer from the object.

2. The method of claim 1, wherein the surface comprises at least one contaminant and wherein, in the passing step (a) the surface of the object is electrolytically stripped of the at least one contaminant.

3. The method of claim 1 wherein the gel-like material comprises an electrolytic agent, a latex formulation, and a chelating agent.

4. The method of claim 3, wherein the electrolytic agent is at least one of a halogen and a transition metal.

5. The method of claim 4, wherein the electrolytic agent is a mixture of iodine, iodide and tri-iodide.

6. The method of claim 3, wherein latex formulation comprises a surfactant, initiator, and at least one monomer.

7. The method of claim 6, wherein, in the curing step (c), the initiator is decomposed and causes polymerization of the at least one monomer.

8. The method of claim 6, wherein the at least one monomer is a vinyl monomer and an acrylic monomer.

9. The method of claim 3, wherein the chelating agent is selected from the group consisting of ethylene-diaminetetraacetic acid (EDTA), nitrilotriacetic acid, ethyleneglycol-bis-(β -aminoethyl ether)-N,N-tetraacetic acid, ethylenediamine, porphine, dimercaprol, and mixtures thereof.

10. The method of claim 1, wherein the uncured gel-like material has a viscosity ranging from about 1,000 to about 100,000 cPs and sufficient conductivity to provide current densities at the contaminated surface of from about 0.5 to about 5 amps/inch².

11. The method of claim 1, wherein the cured gel-like material has a tensile strength ranging from about 200 to about 400 psi.

12. The method of claim 1, wherein, in the applying step (a), a layer of the gel-like material is applied to the surface and further comprising after the passing step (b) and before the curing step (c):

applying additional gel-like material to the surface, wherein the additional gel-like material is free of electric current before the curing step (c).

13. The method of claim 2, wherein, during the passing step (b), a decontaminating apparatus applies a voltage to the gel-like material and wherein the contaminant is a radioactive isotope.

14. The method of claim 13, wherein the decontaminating apparatus comprises a conductive shoe, a porous and permeable abrasive pad through which the gel-like material is applied to the surface, and an insulating standoff to maintain a selected distance between the conductive shoe and the surface.

15. The method of claim 1, wherein, in the passing step (b), a cyclic voltage waveform is applied to the surface.

16. A gel-like material for decontaminating a surface, comprising:

- (a) an electrolytic agent;
- (b) a latex formulation; and
- (c) a chelating agent.

17. The gel-like material of claim 16, wherein the electrolytic agent is at least one of a halogen and a transition metal.

18. The gel-like material of claim 17, wherein the electrolytic agent is a mixture of iodine, iodide and tri-iodide.

19. The gel-like material of claim 16, wherein latex formulation comprises a surfactant, initiator, and at least one monomer.

20. The gel-like material of claim 19, wherein the at least one monomer is selected from the group consisting of rubbers, esters, nylons, acrylates, acetates, butadienes, acrylonitriles, amides, carbonates, acrylics, vinyls, and mixtures thereof.

21. The gel-like material of claim 19, wherein the at least one monomer is a vinyl monomer and an acrylic monomer.

22. The gel-like material of claim 16, wherein the chelating agent is selected from the group consisting of ethylenediaminetetraacetic acid (EDTA), nitrilotriacetic acid, ethyleneglycol-bis-(β -aminoethyl ether)-N,N-tetraacetic acid ethylenediamine, porphine, dimercaprol, and mixtures thereof.

23. The gel-like material of claim 16, wherein, before curing, gel-like material has a viscosity ranging from about 1,000 to about 100,000 cPs and sufficient conductivity to provide current densities at the contaminated surface of from about 0.5 to about 5 amps/inch².

24. The gel-like material of claim 16, wherein, after curing, the gel-like material has a tensile strength ranging from about 200 to about 400 psi.

25. A decontaminating system, comprising:

a decontaminating apparatus comprising:

- an electrically conductive scrubbing shoe having an inlet for a gel-like material; and
- an insulating standoff positioned between the scrubbing shoe and a surface to be decontaminated to maintain a desired distance between the scrubbing shoe and the surface;

a reservoir for the gel-like material, the reservoir being in communication with the inlet; and

a voltage source in communication with the electrically conductive scrubbing shoe, whereby a current is passed through the gel-like material applied to the surface, thereby removing contaminants from the surface.

26. The decontaminating system of claim 25, wherein the decontaminating apparatus further comprises an abrasive pad positioned between the scrubbing shoe and the surface.

27. The decontaminating system of claim 26, wherein the abrasive pad is porous and permeable to permit the gel-like material to pass from the inlet through the pad and onto the surface.

28. The decontaminating system of claim 25, further comprising:

a processor and power control module operable to cause the voltage source to apply a cyclic voltage waveform to the shoe.

29. The decontaminating system of claim 28, further comprising:

a device for measuring at least one of the voltage applied to the shoe and the current and wherein the processor is operable, based on the measured at least one of the voltage and current, to account for drift in the applied voltage waveform.

30. The decontaminating system of claim 25, further comprising:

a memory operable to contain a plurality of differing voltage waveforms for a plurality of differing types of surfaces to be contaminated; and

a processor operable to select a voltage waveform corresponding to a selected type of surface to be contaminated and cause application of the selected voltage waveform to the surface.

31. The decontaminating system of claim 25, wherein the scrubbing shoe comprises a trowell edge to manipulate the gel-like material on the surface.

32. An arrangement for decontaminating a surface, comprising:

a contaminated surface;

a layer of a gel-like material applied to the contaminated surface; and

an electrically conductive surface in contact with a free surface of the gel-like material, the electrically conductive surface being in electrical communication with a power source to pass an electric current through the gel-like material and contaminated surface, wherein the layer is positioned between the contaminated surface and the electrically conductive surface.

33. The arrangement of claim 32, wherein the gel-like material comprises an electrolytic agent, a latex formulation, and a chelating agent.

34. The arrangement of claim 33, wherein the electrolytic agent is at least one of a halogen and a transition metal.

35. The arrangement of claim 34, wherein the electrolytic agent is a mixture of iodine, iodide and tri-iodide.

36. The arrangement of claim 34, wherein latex formulation comprises a surfactant, initiator, and at least one monomer.

37. The arrangement of claim 36, wherein the initiator is decomposed and causes polymerization of at least one monomer.

38. The arrangement of claim 36, wherein the at least one monomer is a vinyl monomer and an acrylic monomer.

39. The arrangement of claim 33, wherein the chelating agent is selected from the group consisting of ethylenediaminetetraacetic acid (EDTA), nitrilotriacetic acid, ethyleneglycol-bis-(β -aminoethyl ether)-N,N-tetraacetic acid, ethylenediamine, porphine, dimercaprol, and mixtures thereof.

40. The arrangement of claim 32, wherein the uncured gel-like material has a viscosity ranging from about 1,000 to about 100,000 cPs, and sufficient conductivity to provide current densities at the contaminated surface of from about 0.5 to about 5 amps/inch².

41. The arrangement of claim 32, wherein the cured gel-like material has a tensile strength ranging from about 200 to about 400 psi.

42. The arrangement of claim 32, wherein the conductive surface is part of a decontaminating apparatus that comprises a conductive shoe, a porous and permeable abrasive pad through which the gel-like material is applied to the surface, and an insulating standoff to maintain a selected distance between the conductive shoe and the surface and wherein the conductive surface is a surface of the conductive shoe.

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