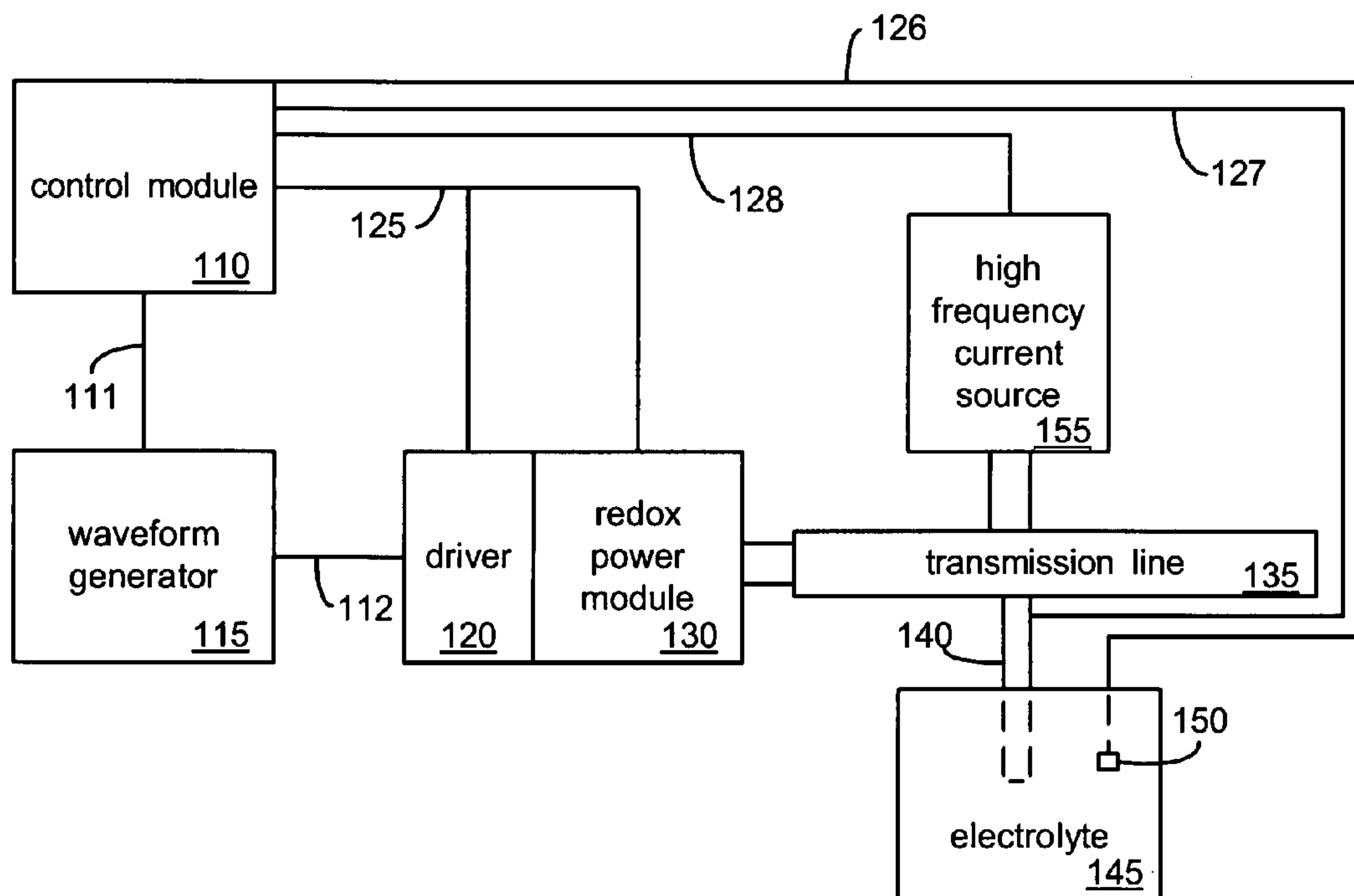


US 20090134039A1

(19) **United States**(12) **Patent Application Publication**
MATTHEWS(10) **Pub. No.: US 2009/0134039 A1**(43) **Pub. Date: May 28, 2009**(54) **SYSTEM AND METHOD FOR ISOTOPE
SELECTIVE CHEMICAL REACTIONS****Publication Classification**(51) **Int. Cl.**
C25C 3/00 (2006.01)(52) **U.S. Cl.** **205/354**(57) **ABSTRACT**(76) Inventor: **MEHLIN DEAN MATTHEWS,**
Saratoga, CA (US)Correspondence Address:
MEHLIN DEAN MATTHEWS
P.O. BOX 3340
ALBANY, OR 97321 (US)(21) Appl. No.: **12/036,282**(22) Filed: **Feb. 24, 2008****Related U.S. Application Data**(60) Provisional application No. 60/990,913, filed on Nov.
28, 2007.

A system providing excitation and selective transport and/or reaction of isotopologues. Two or more isotope effects are combined within a reaction cell to produce transient differentiation of isotopologues through excitation or chemical reaction followed by rapid separation or conversion. A current conductor adjacent to a fluid carries an excitation current produced by a switching source. The excitation current produces an alternating magnetic field adjacent to the conductor that interacts with a population of isotopologues within the adjacent fluid layer to produce a population of reaction intermediates or products that have an isotopic composition different from the initial isotopologues. An electrolytic power supply may also be connected to the conductor to provide a voltage that may be used to selectively attract and electrolyze a portion of the reaction intermediates or products within the adjacent fluid layer.

100

100

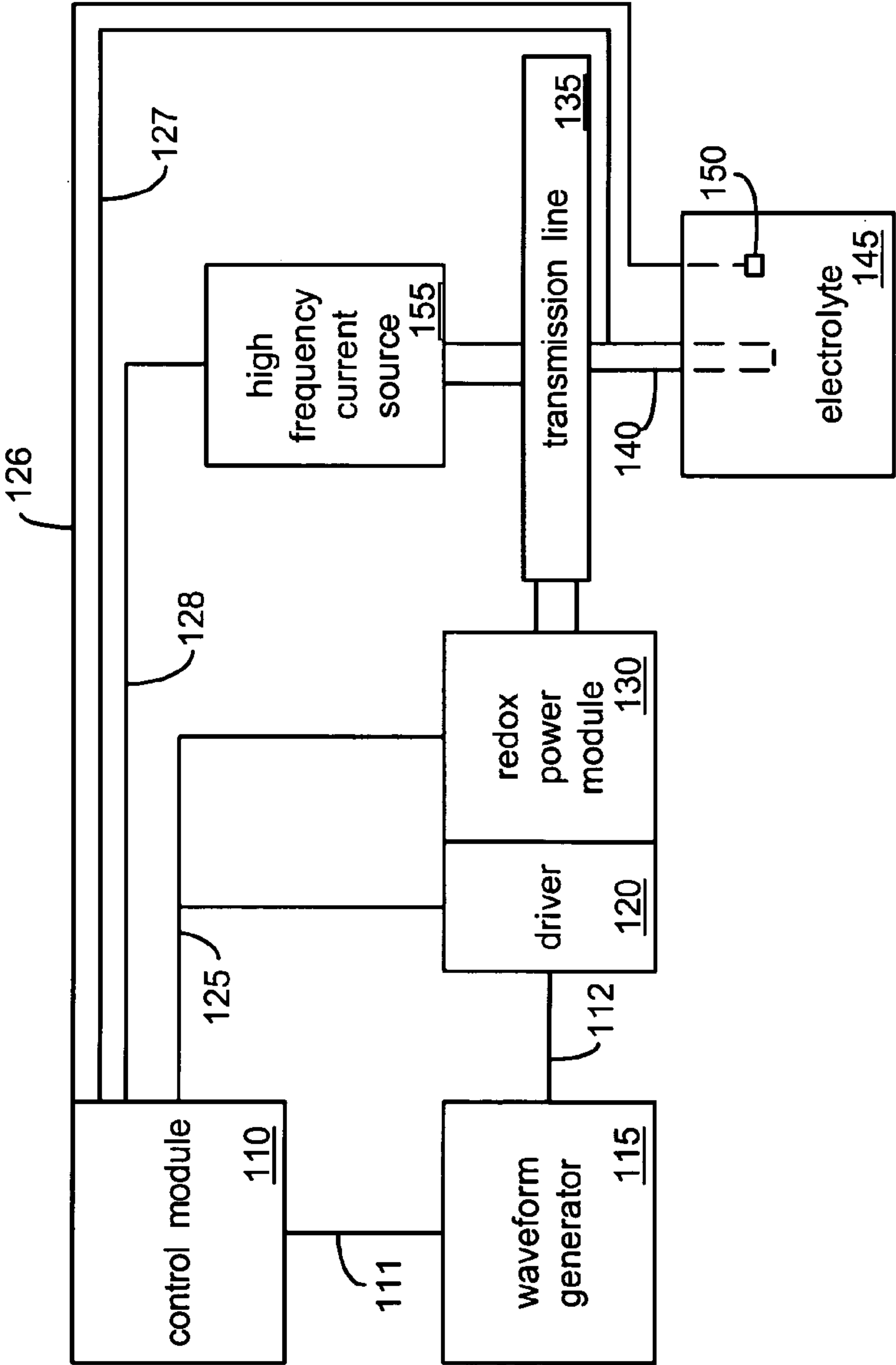


FIG. 1A

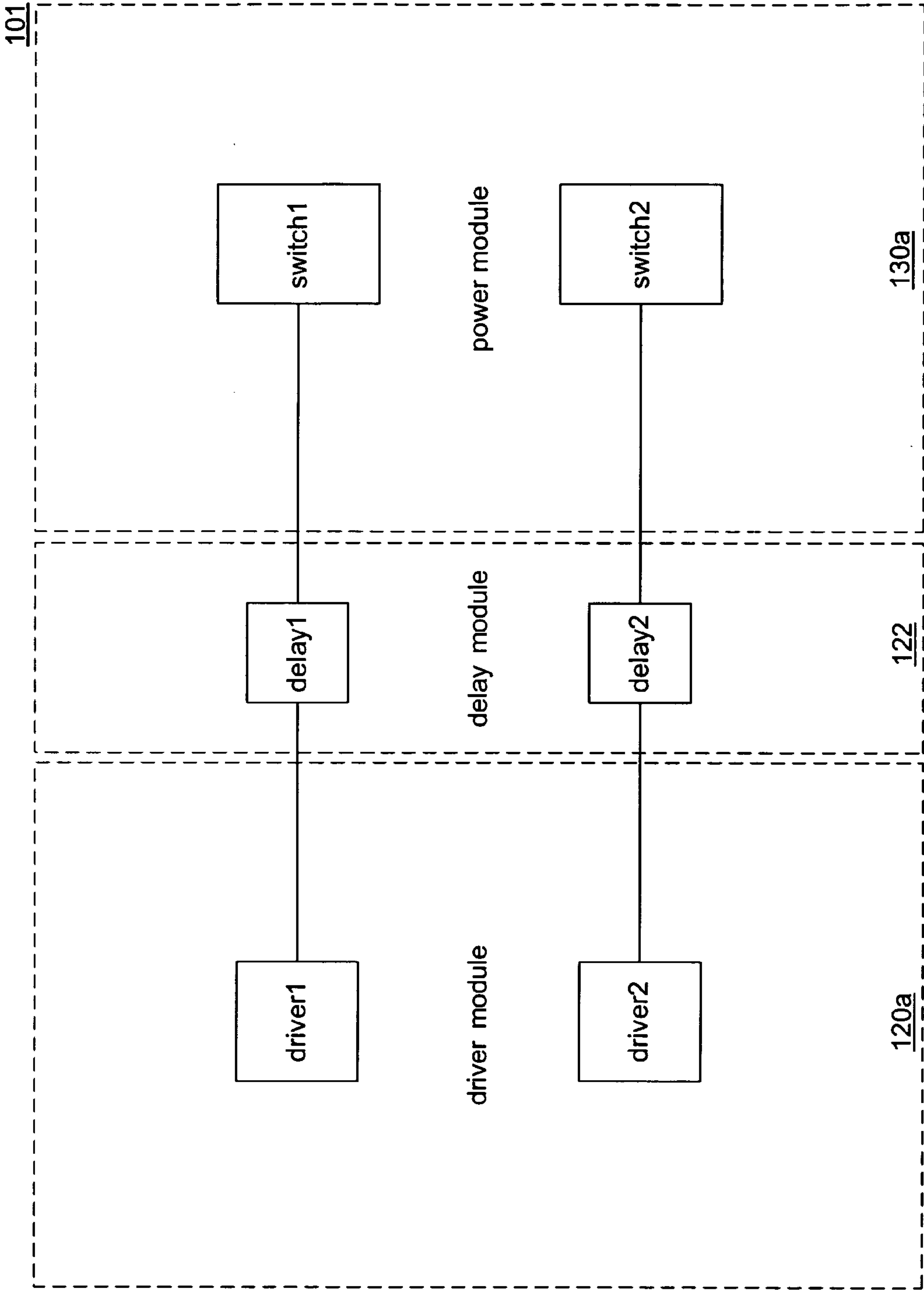


FIG. 1B

200

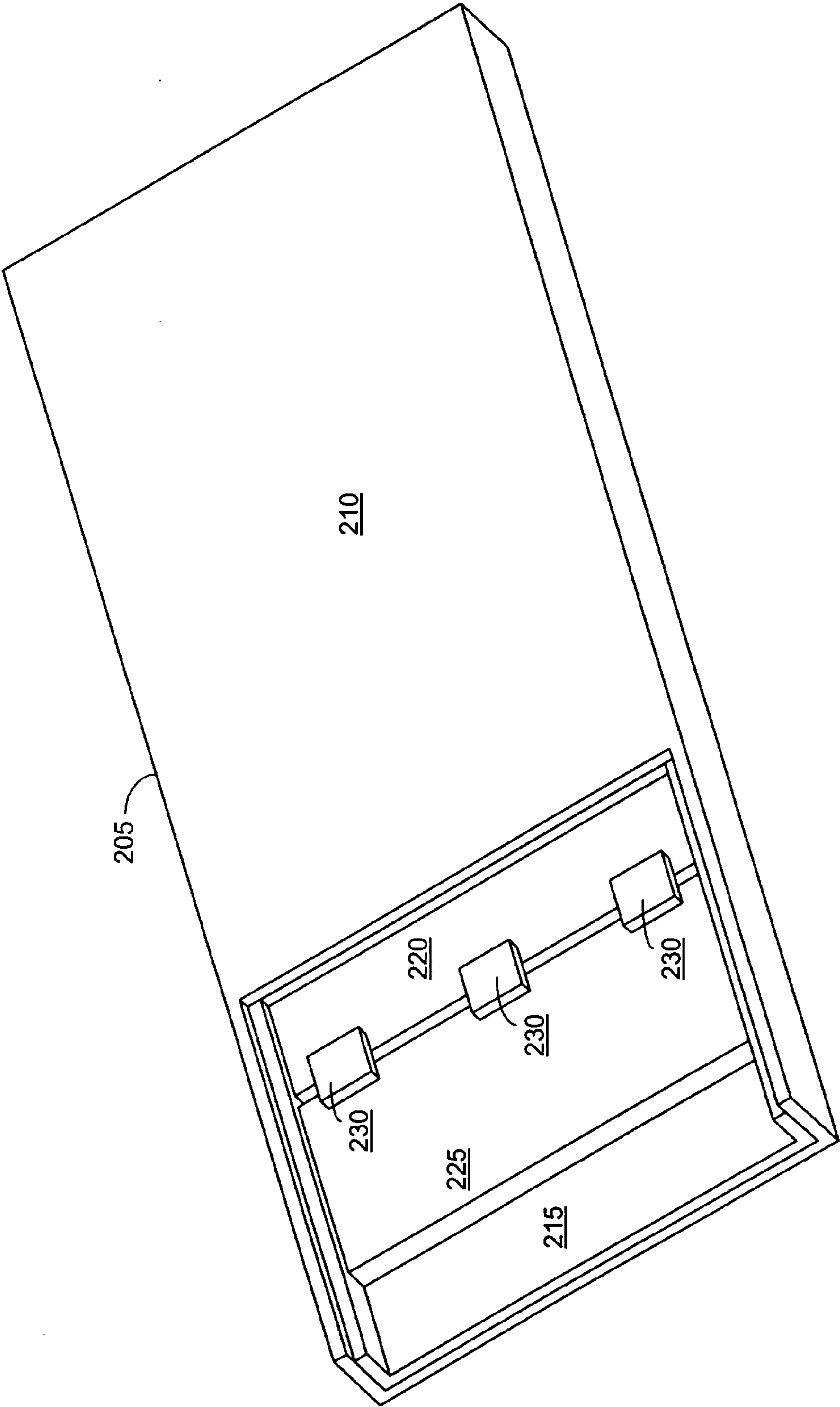


FIG. 2A

201

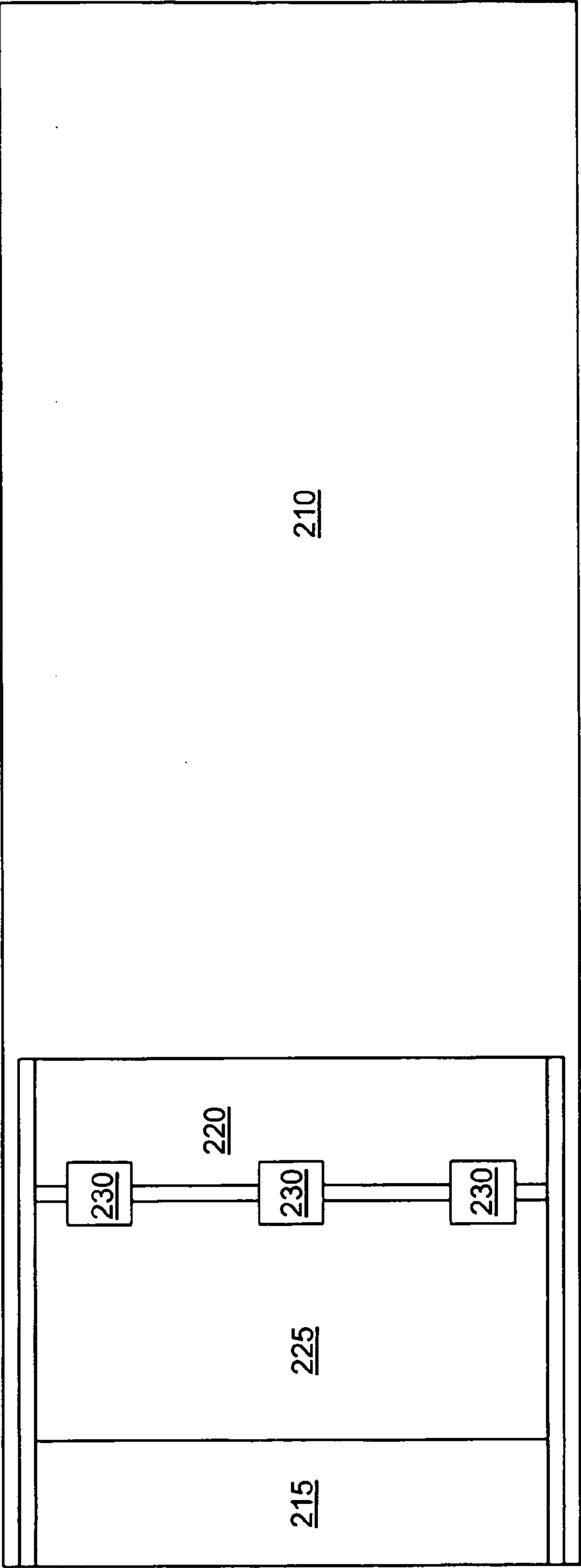


FIG. 2B

202

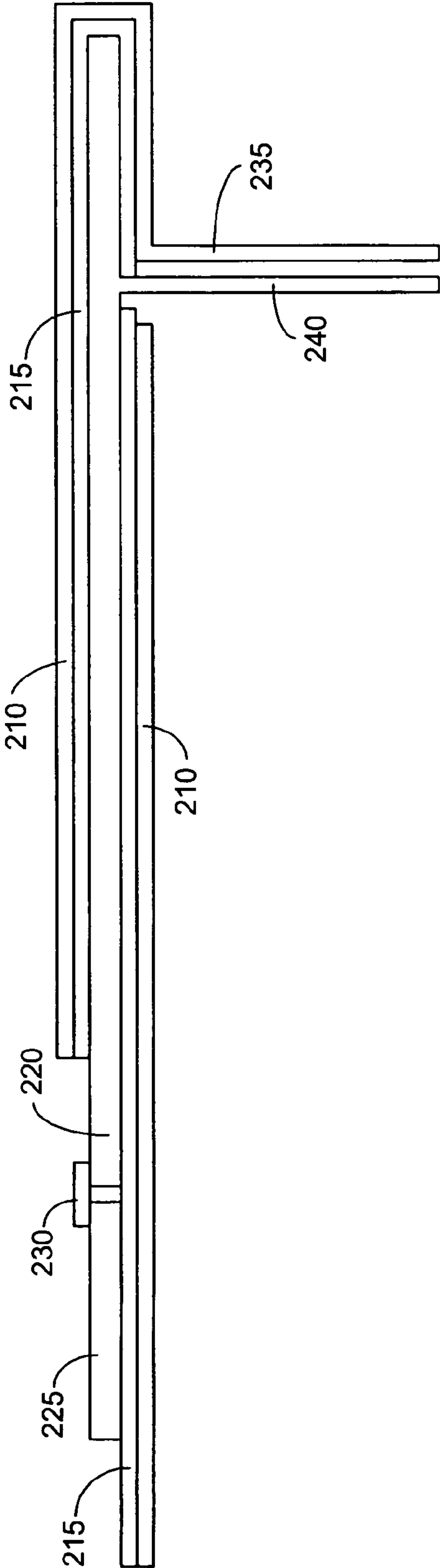


FIG. 2C

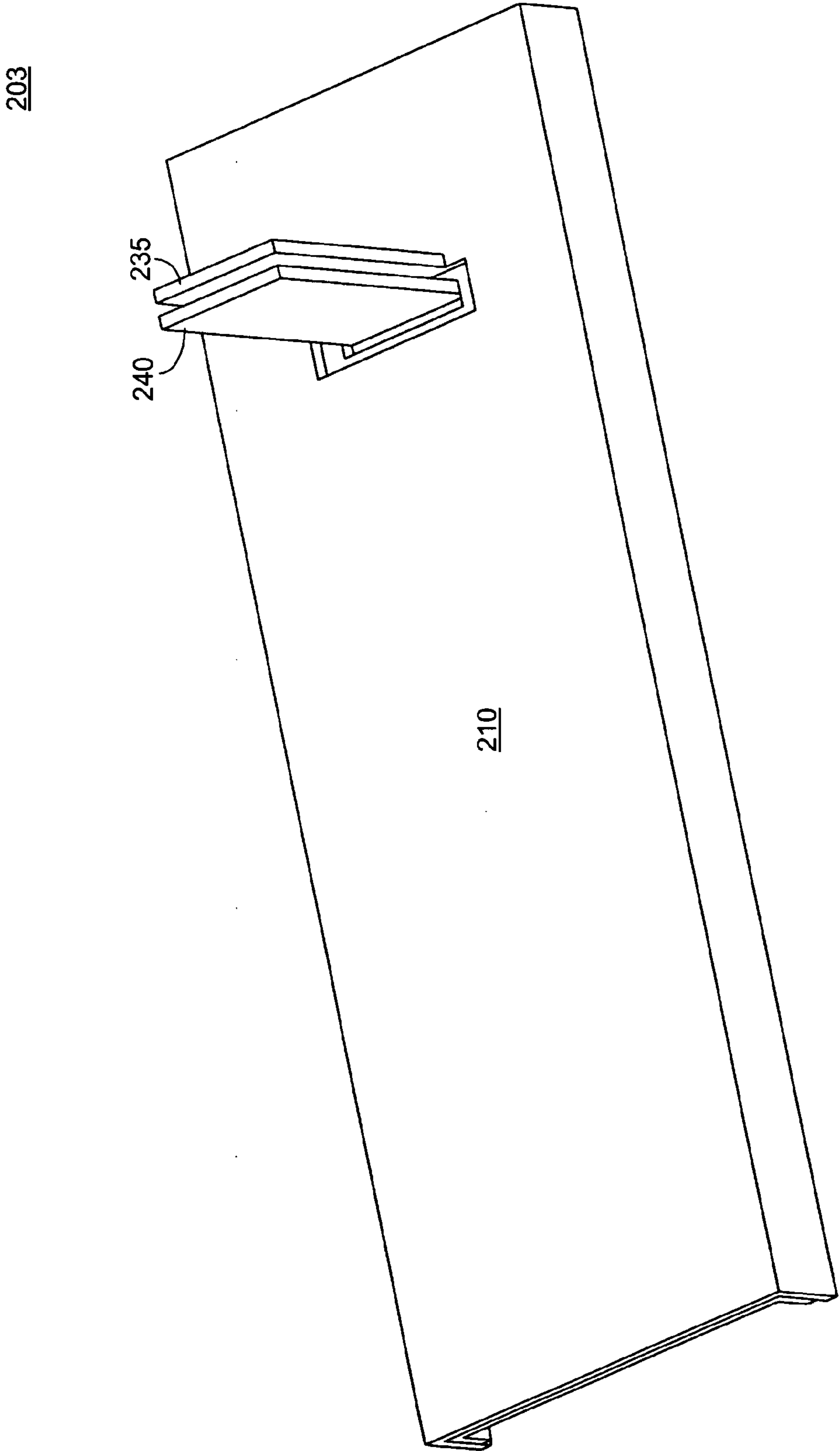


FIG. 2D

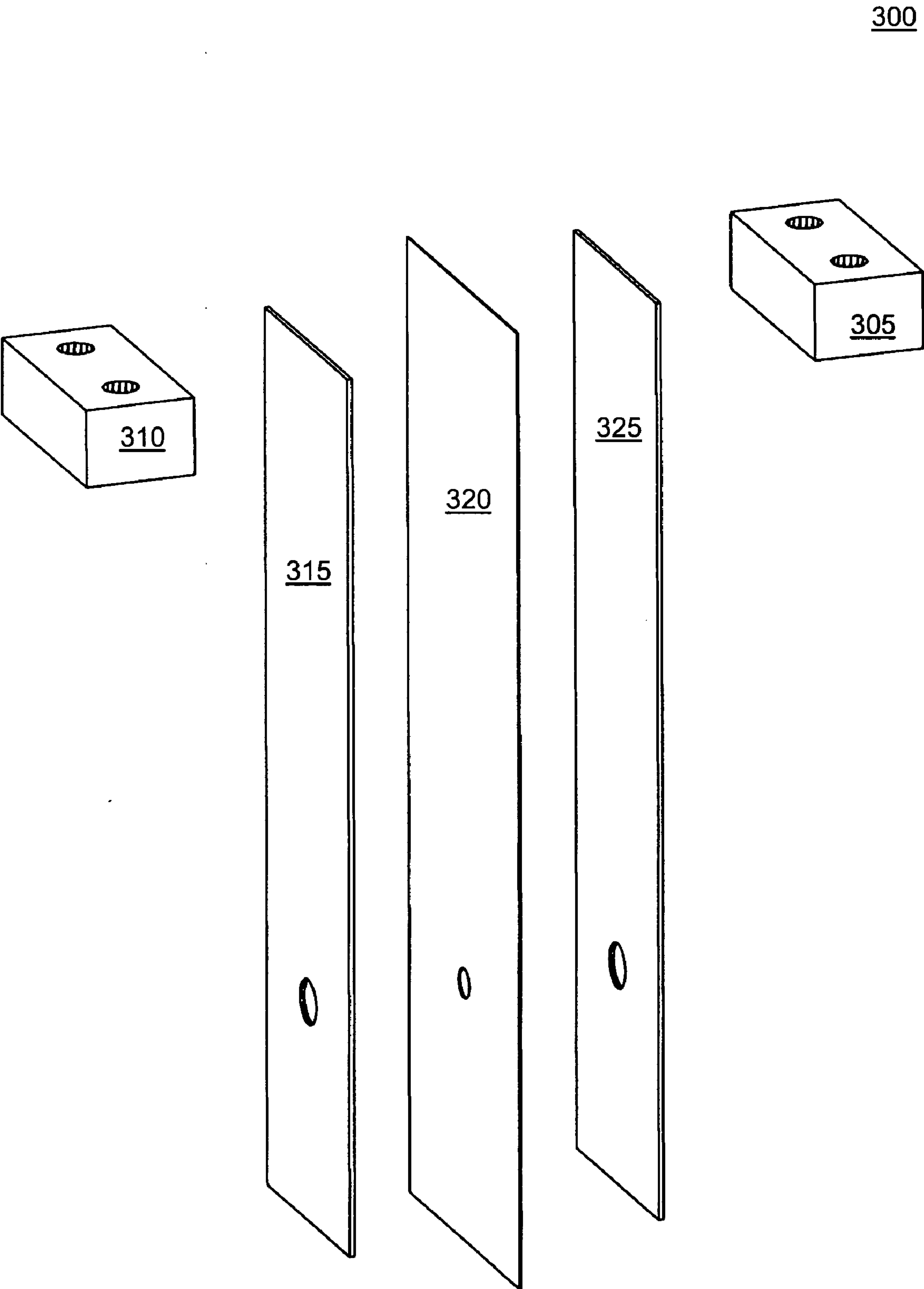


FIG. 3A

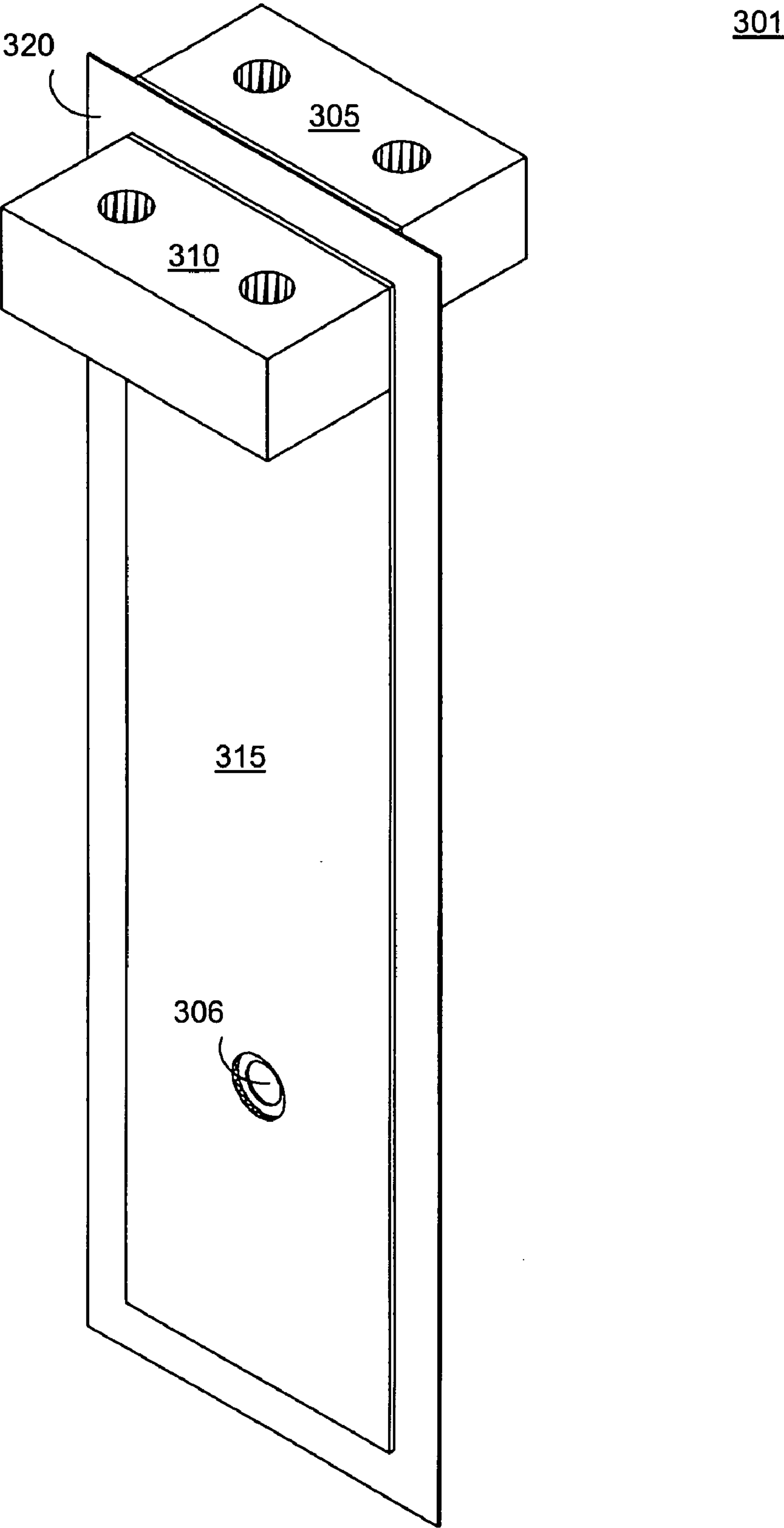


FIG. 3B

302

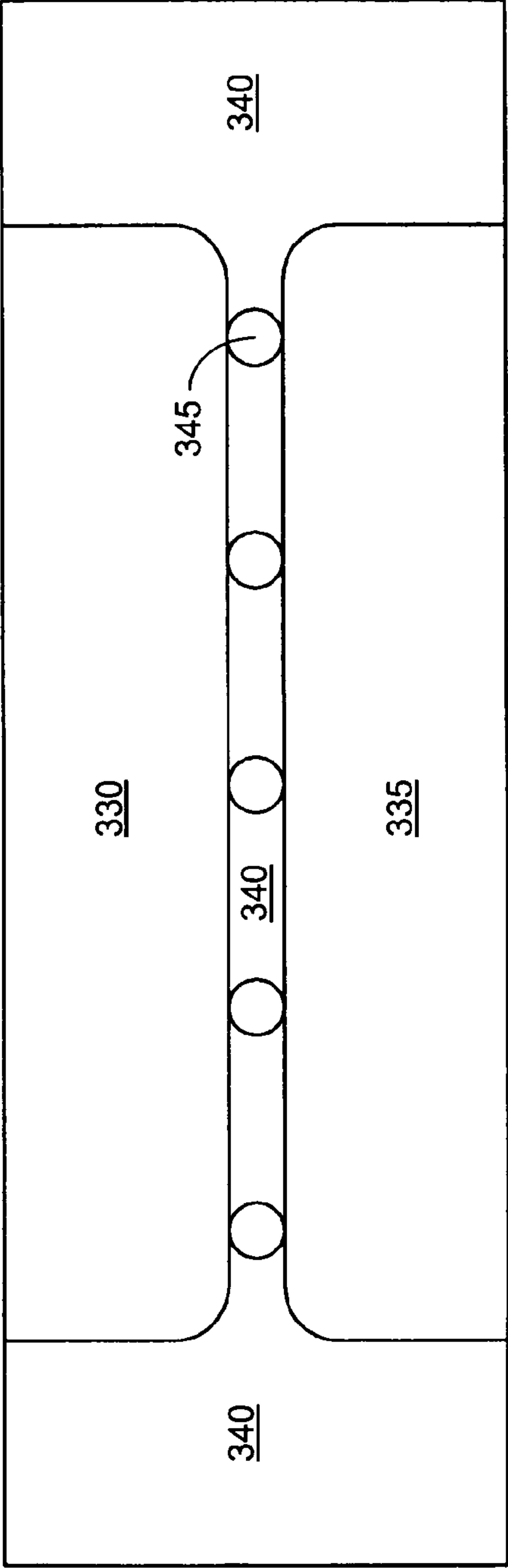


FIG. 3C

400

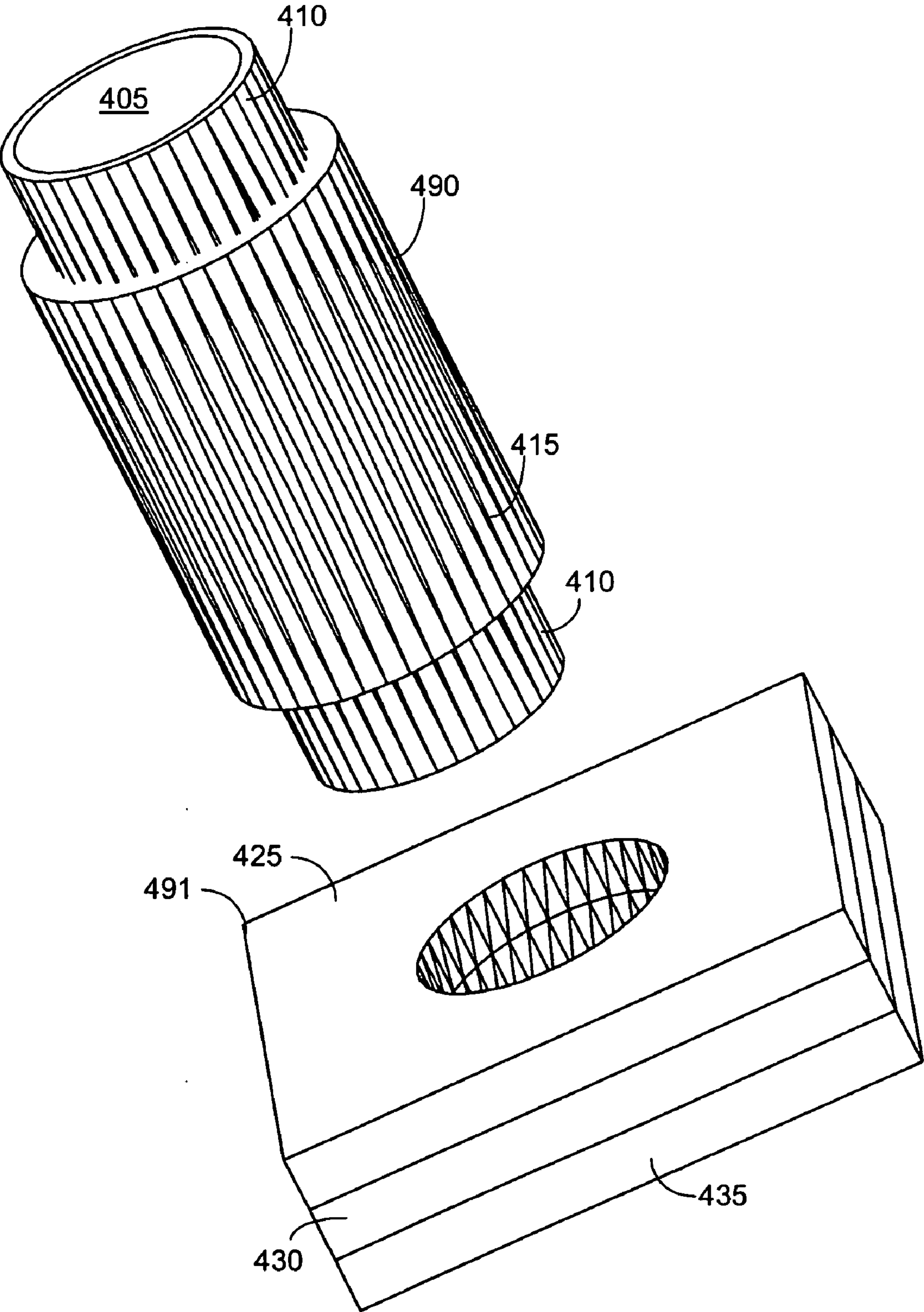


FIG. 4A

401

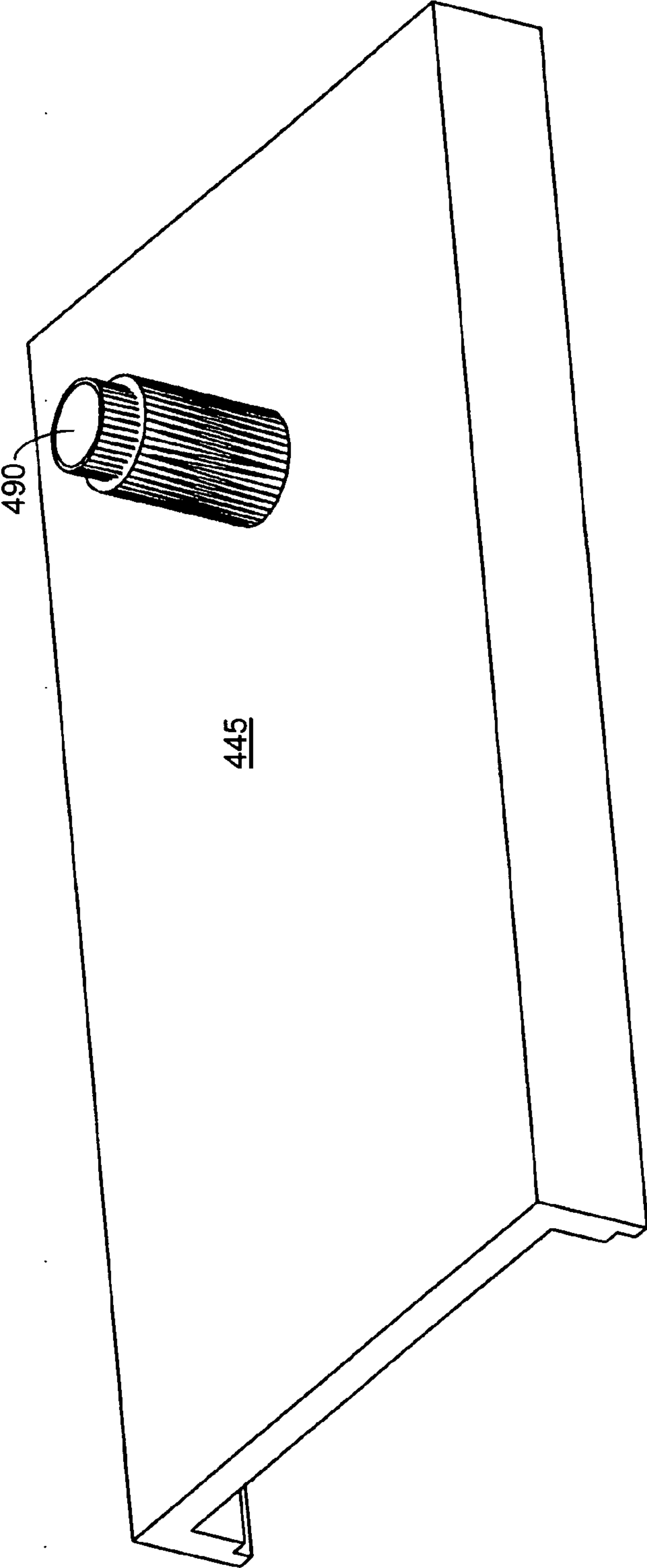
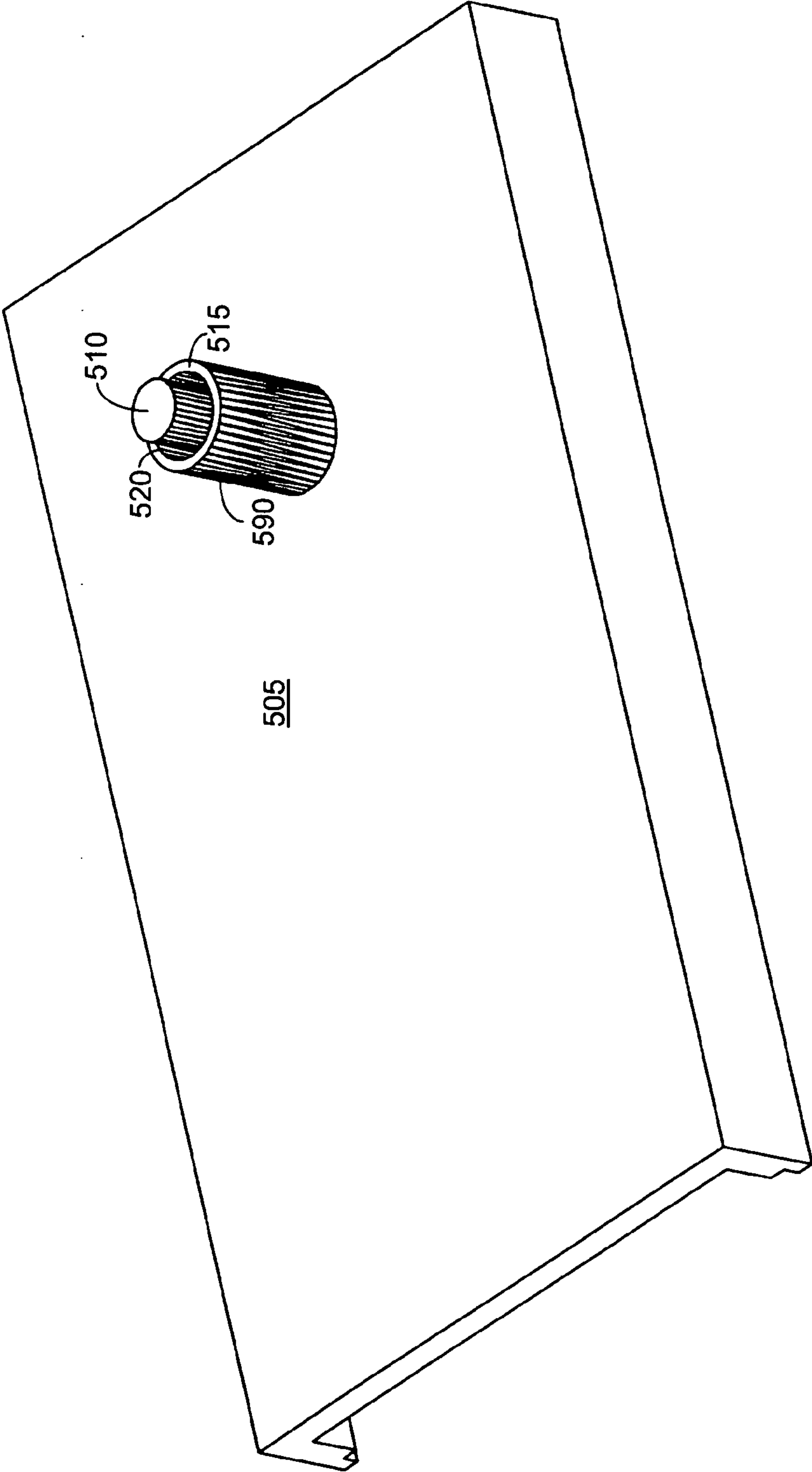


FIG. 4B

500



505

FIG. 5

600

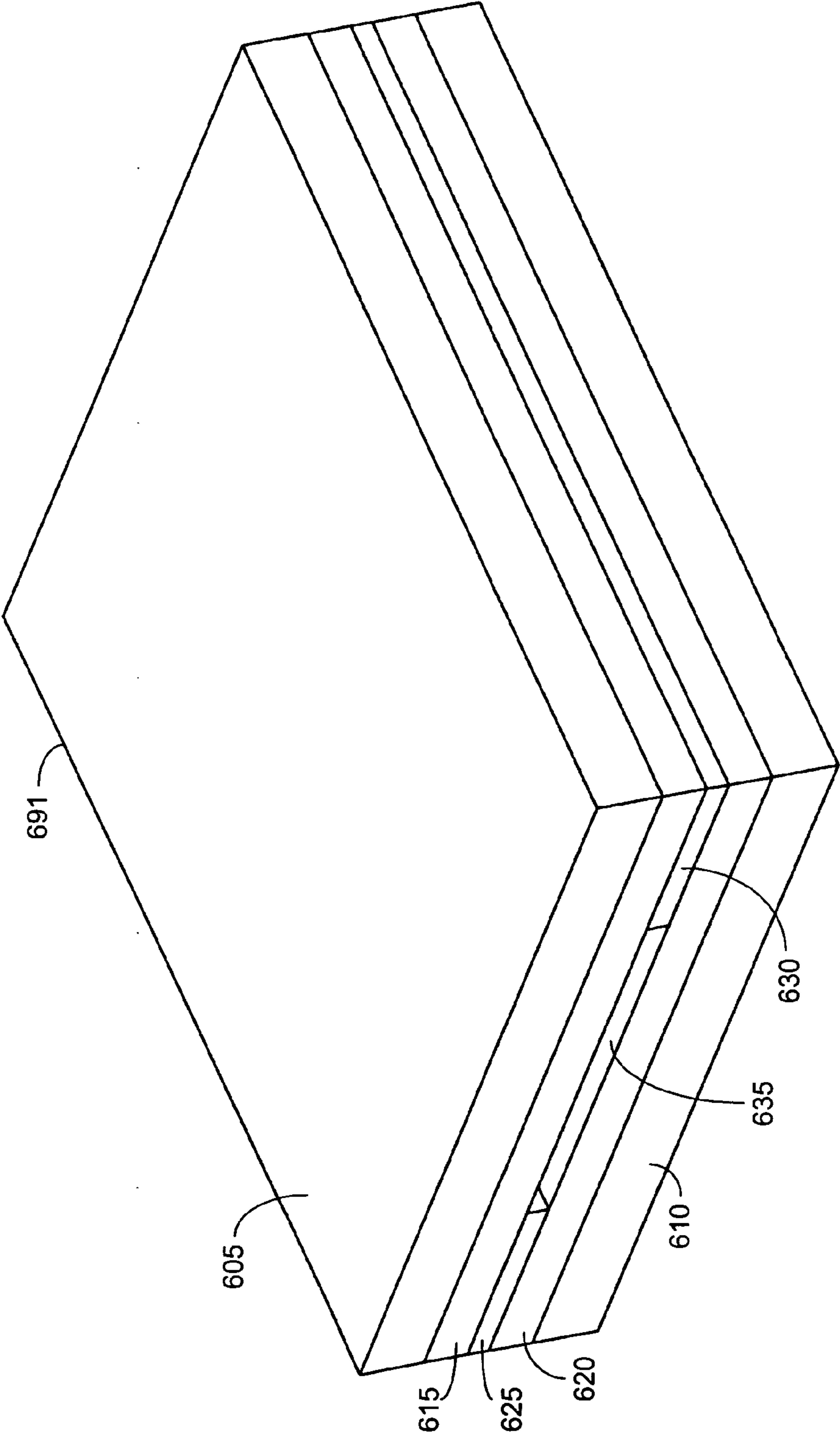


FIG. 6A

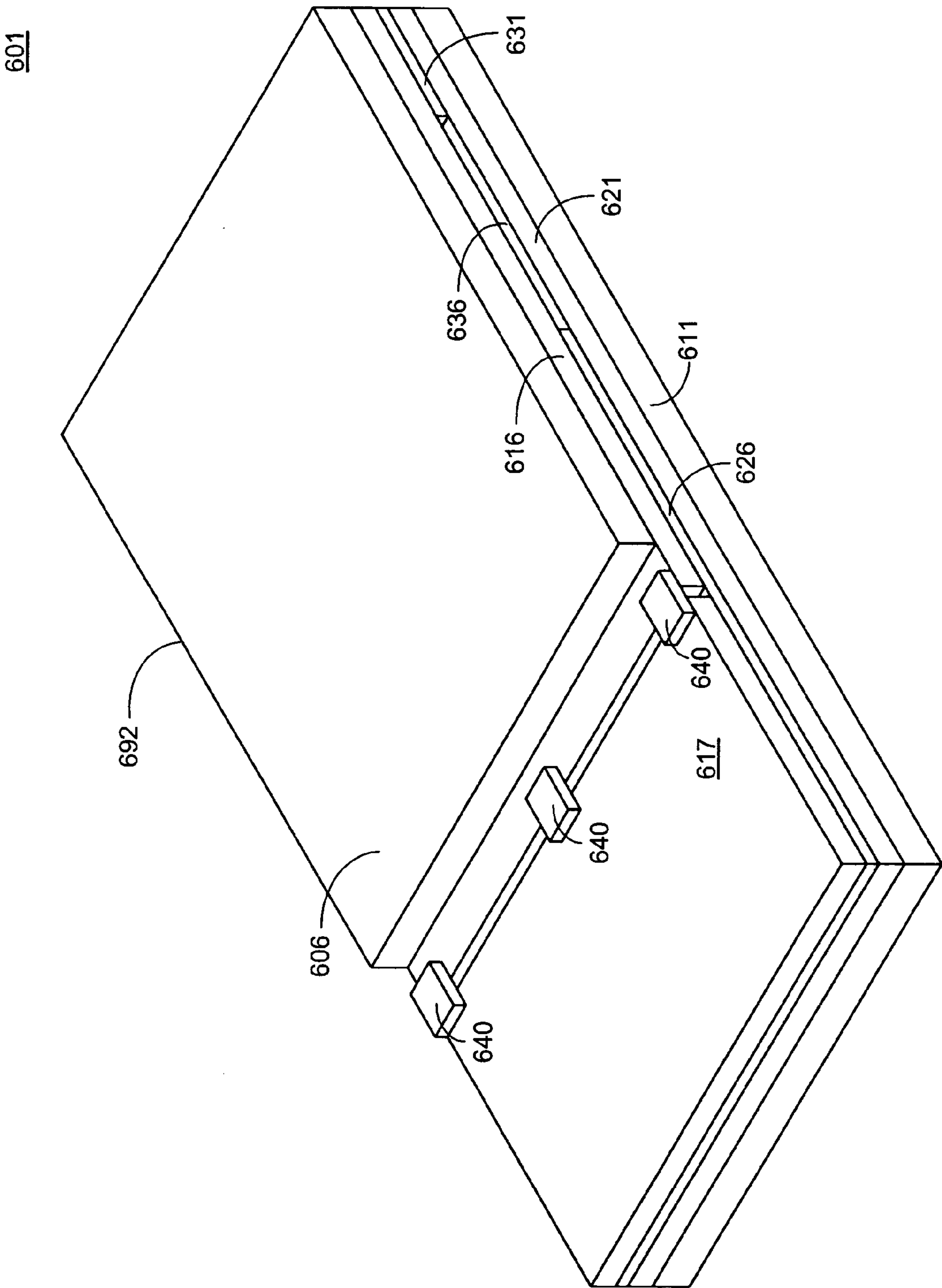


FIG. 6B

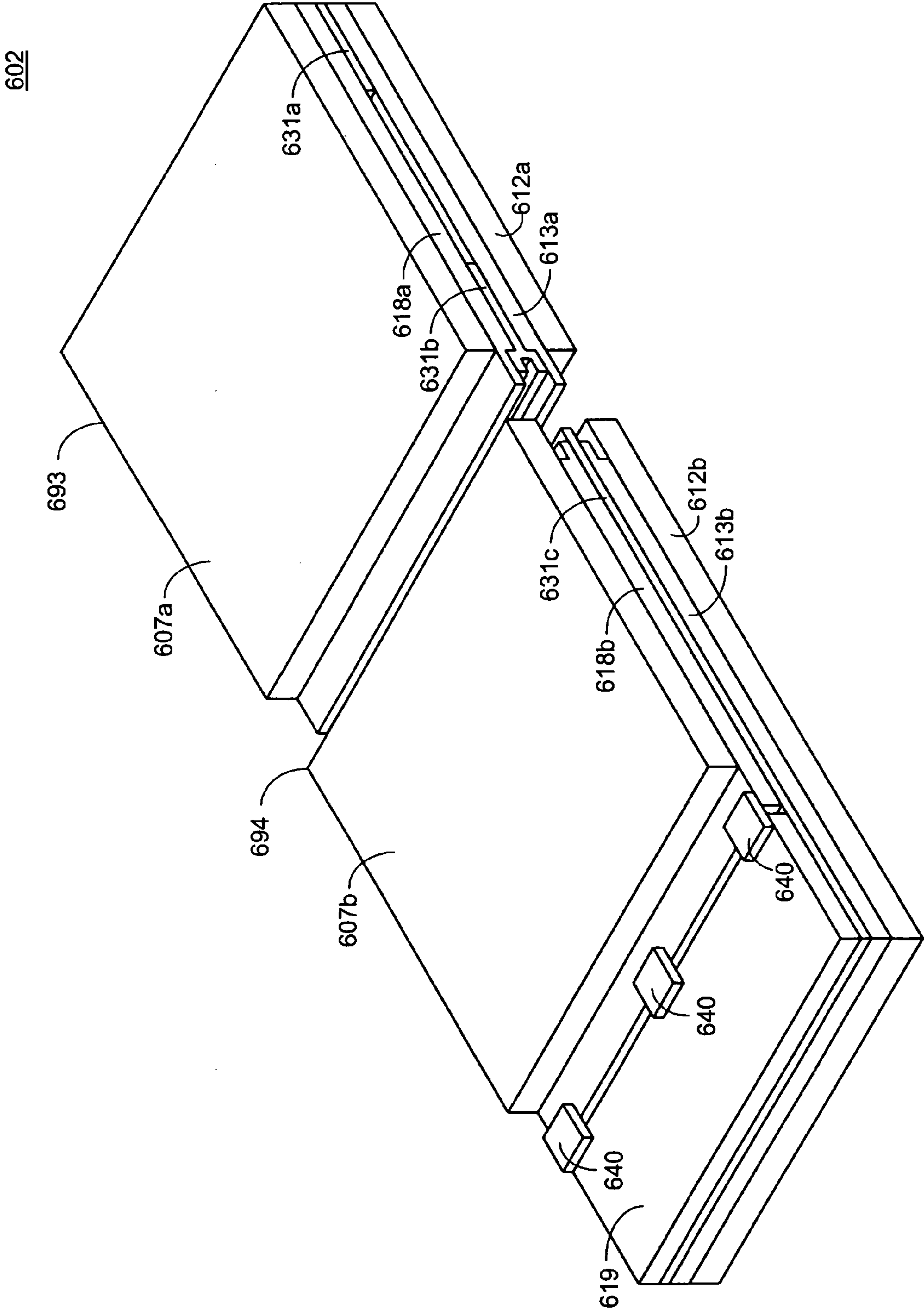


FIG. 6C

603

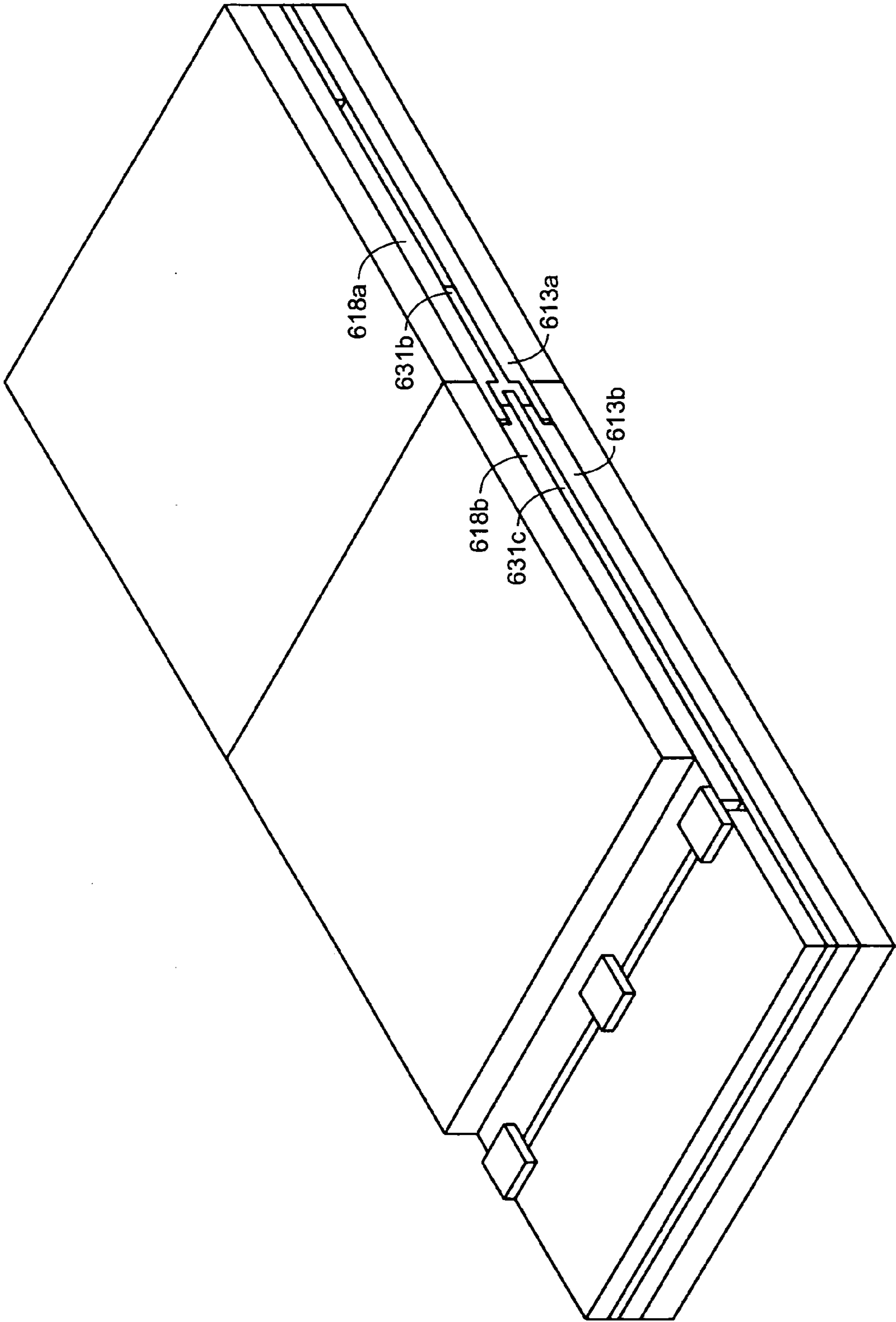


FIG. 6D

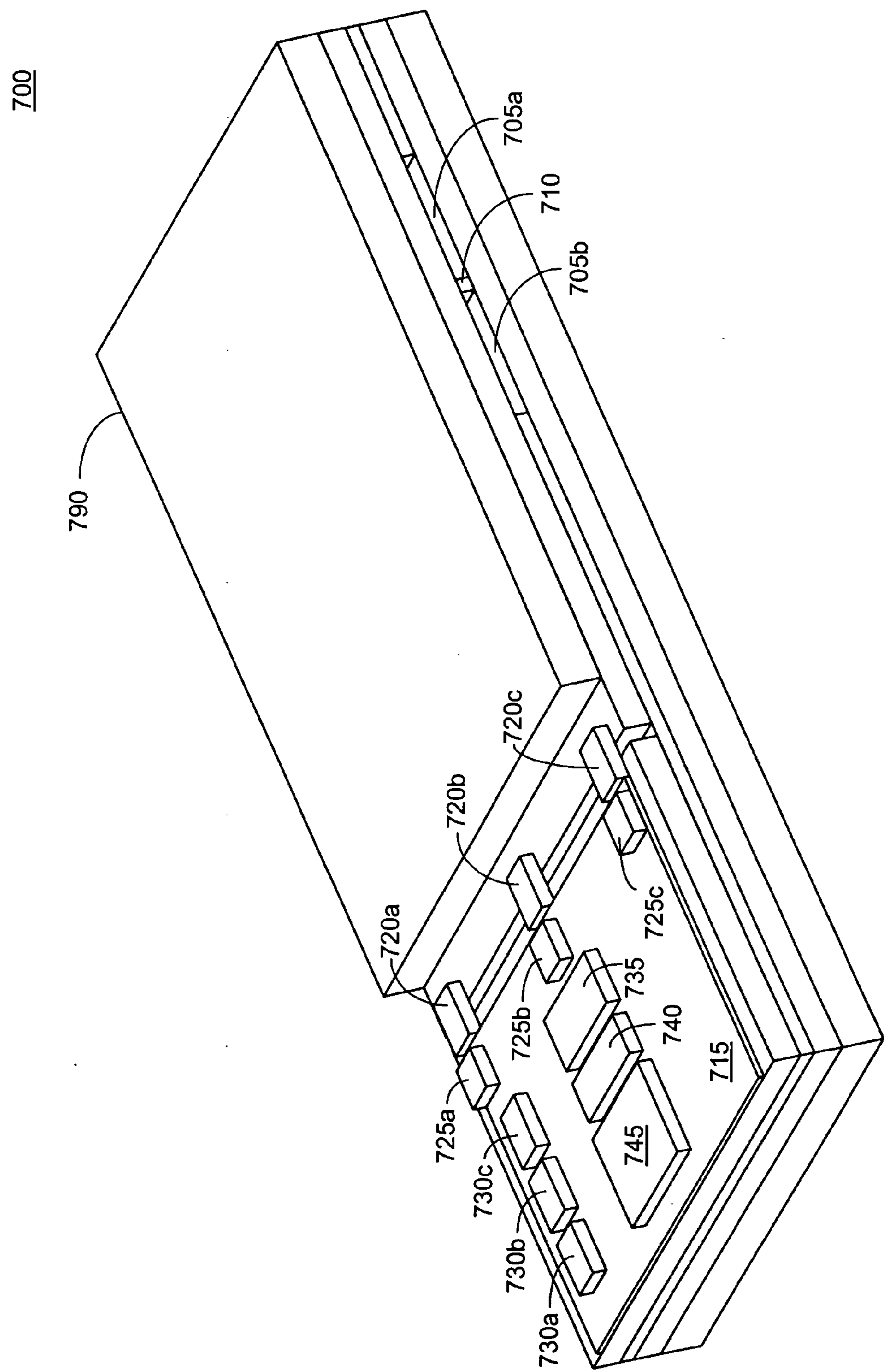


FIG. 7A

701

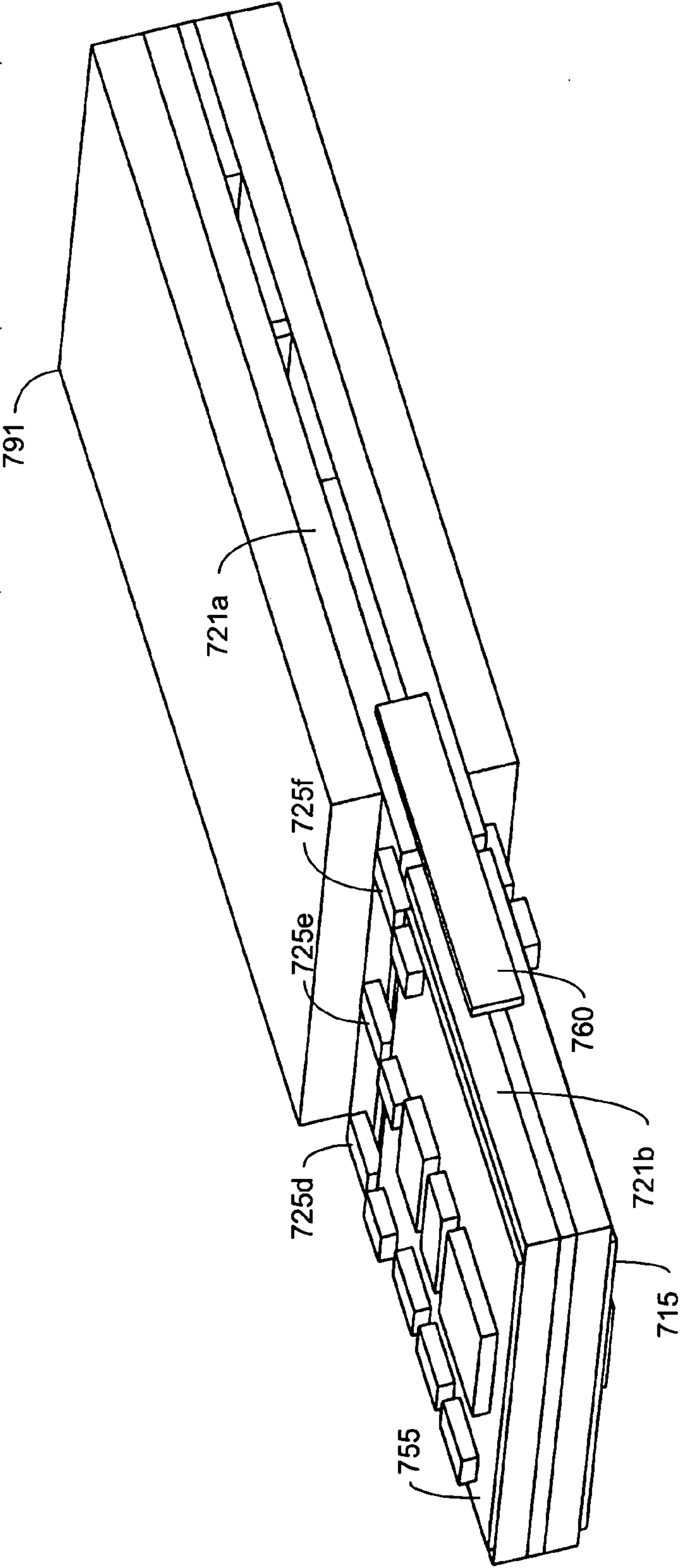


FIG. 7B

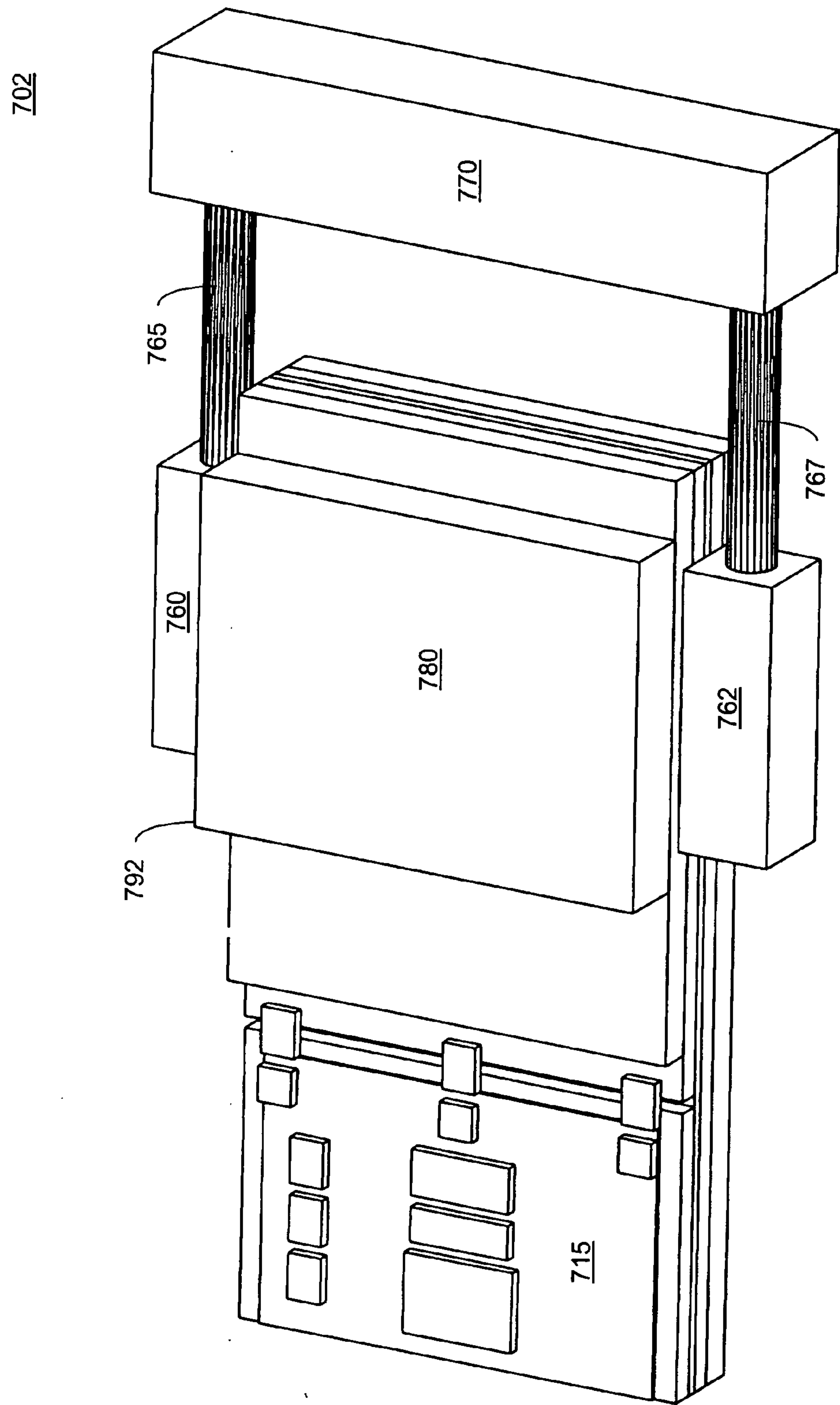


FIG. 7C

800

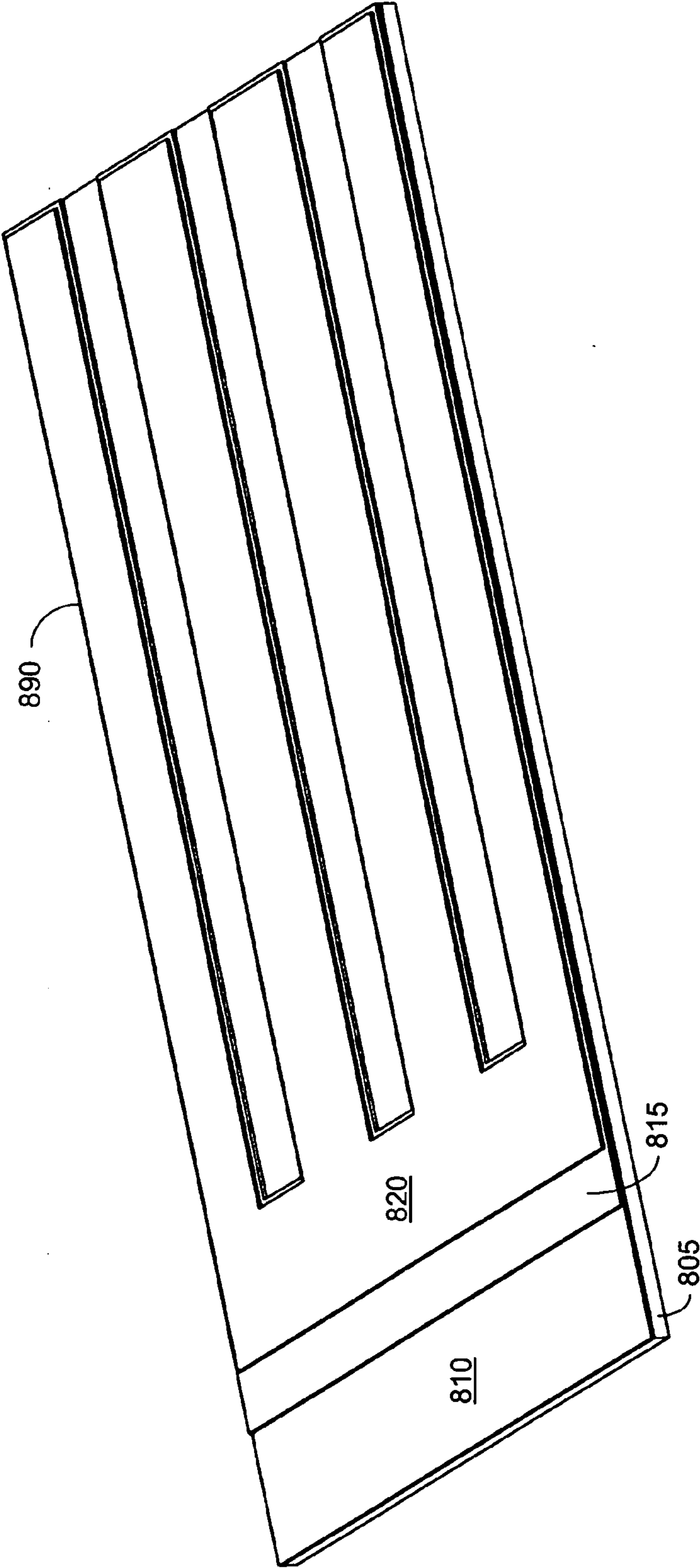


FIG. 8A

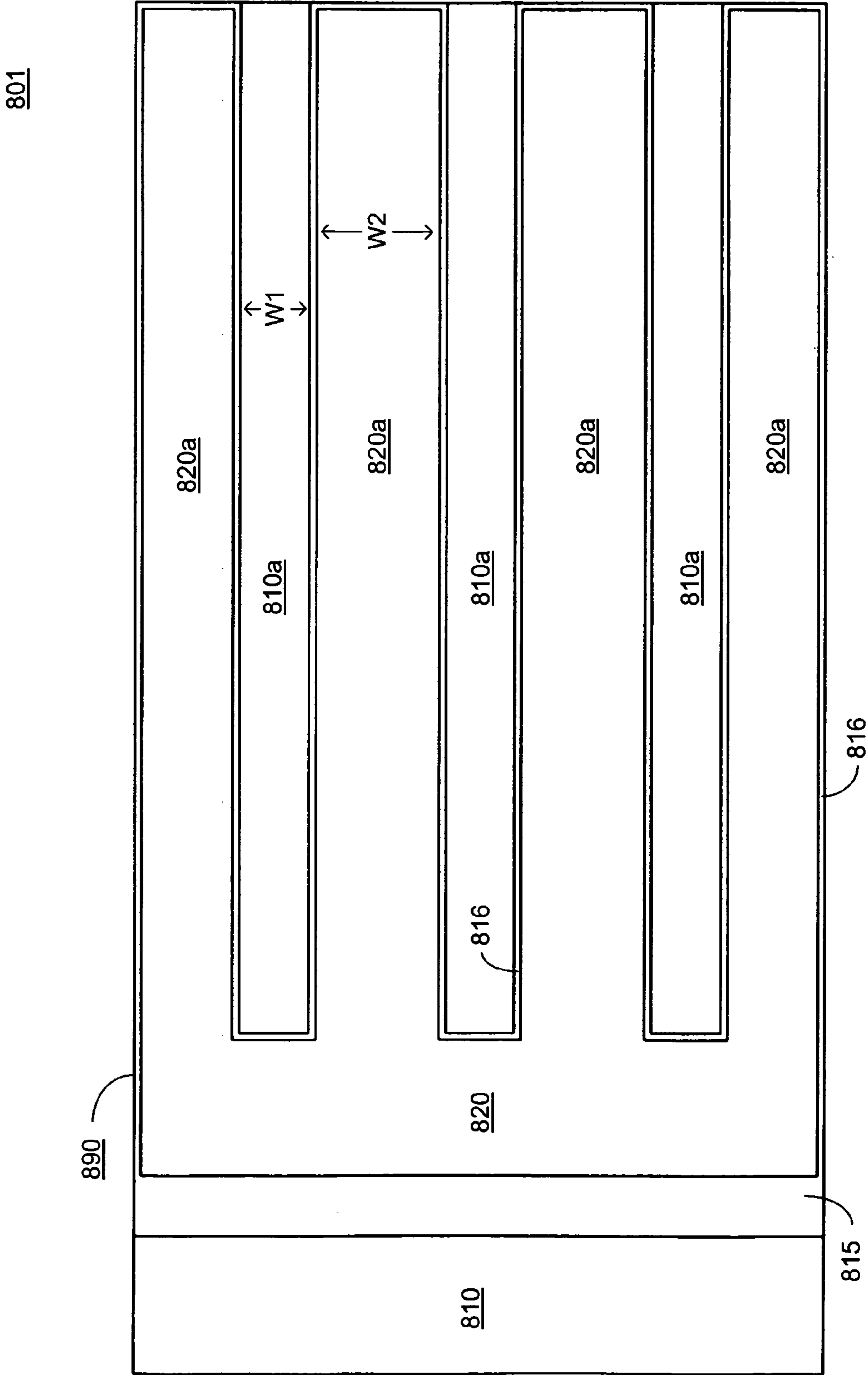


FIG. 8B

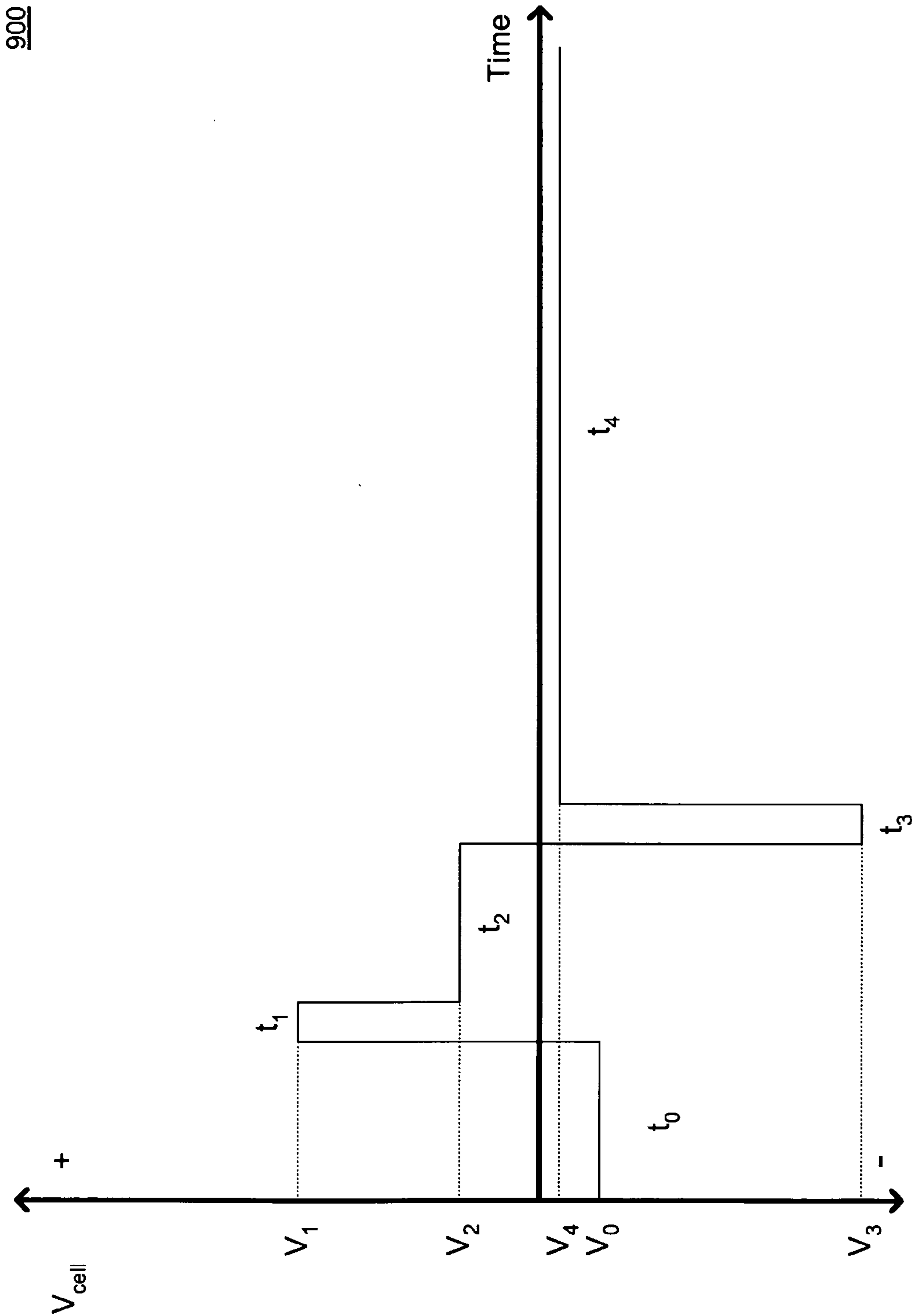


FIG. 9

1000

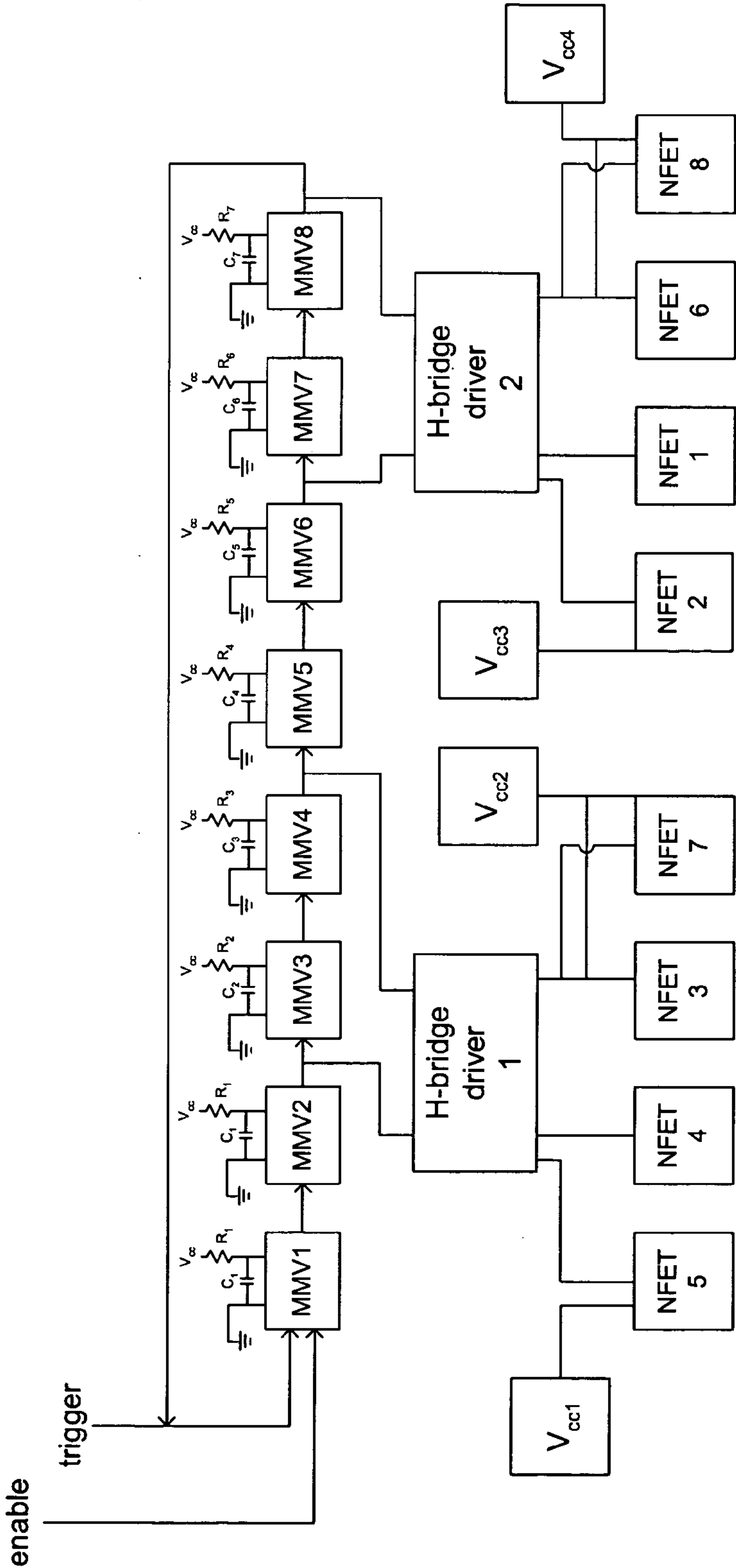


FIG. 10

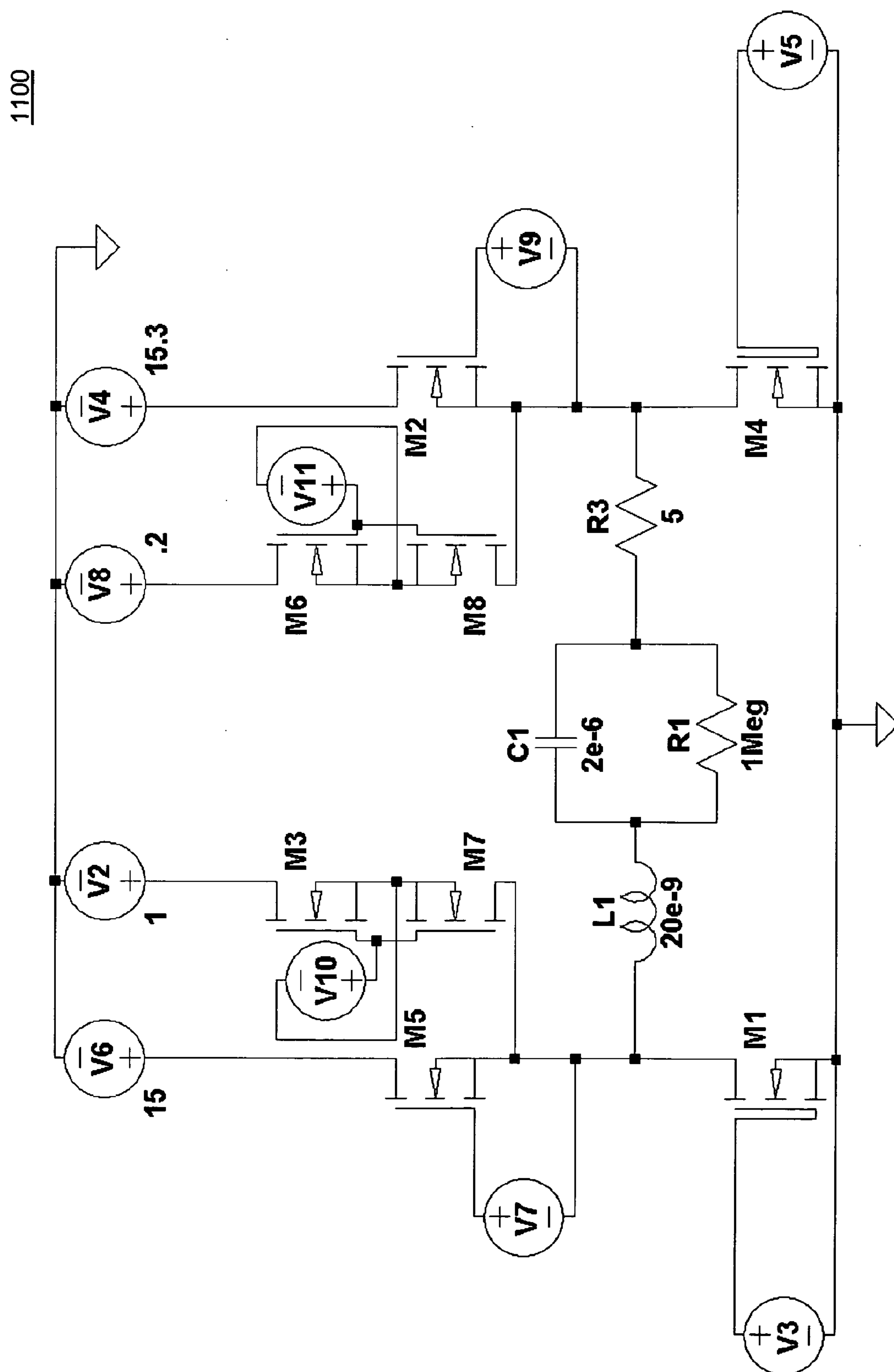


FIG. 11

1200

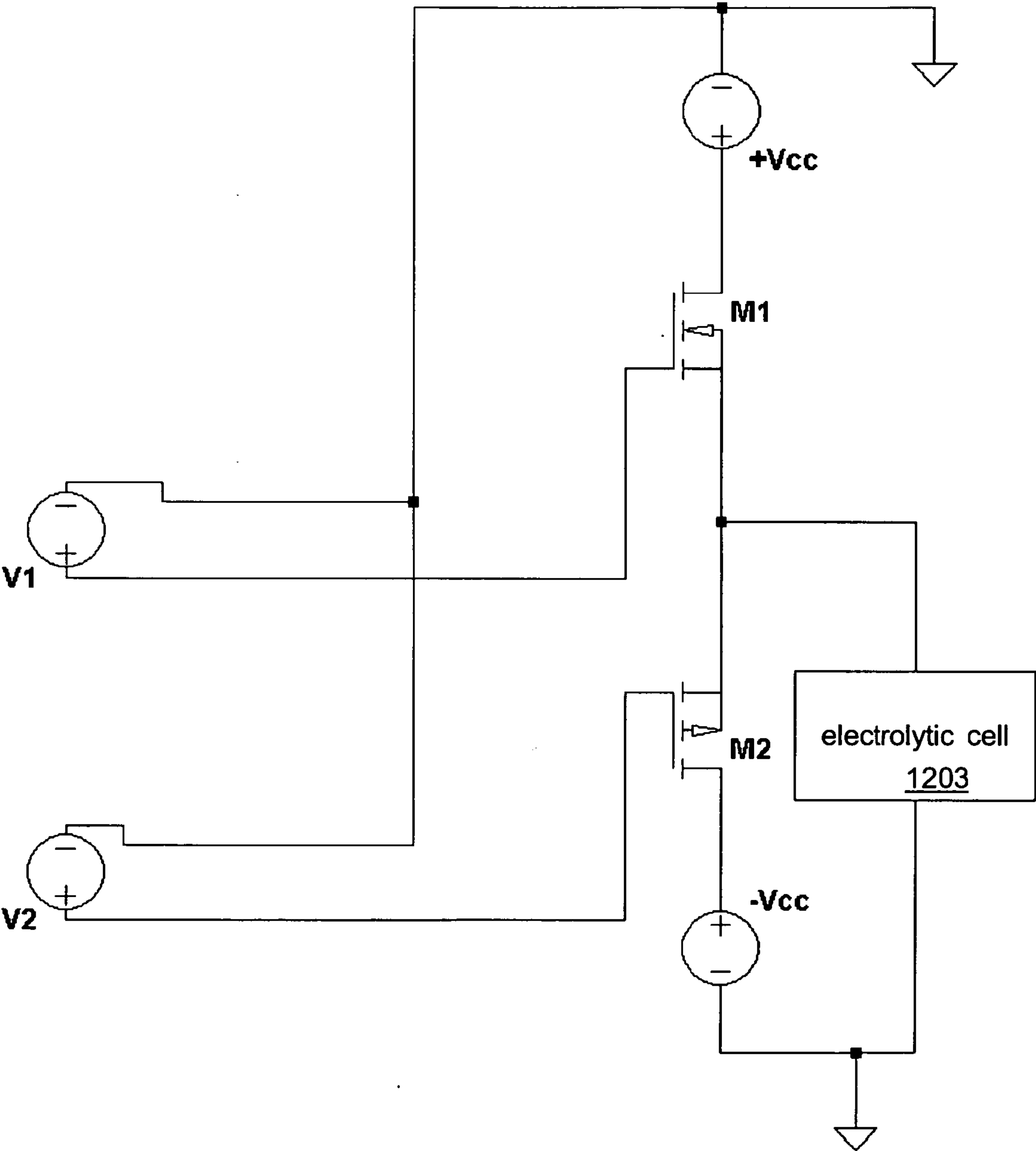


FIG. 12A

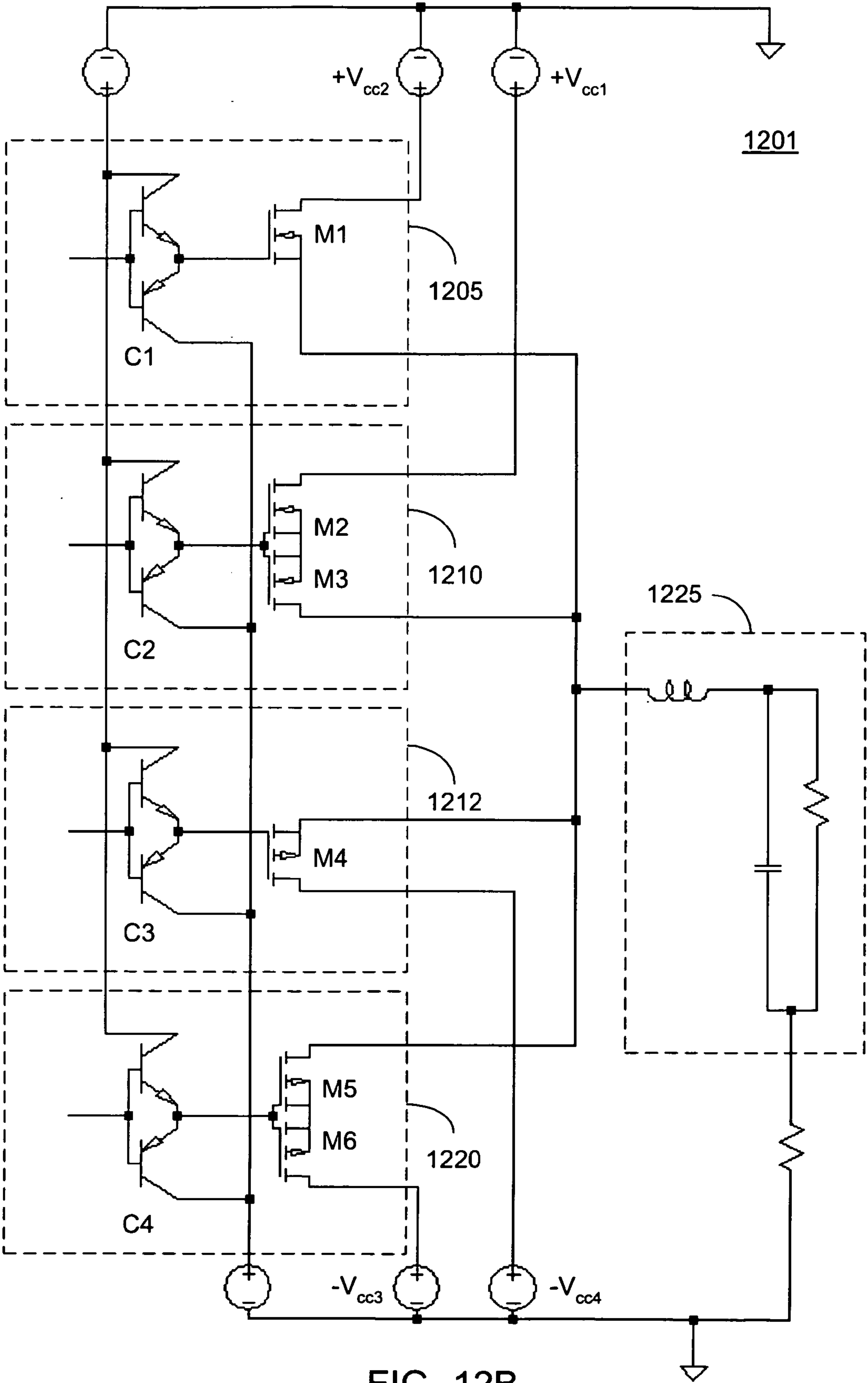


FIG. 12B

1300

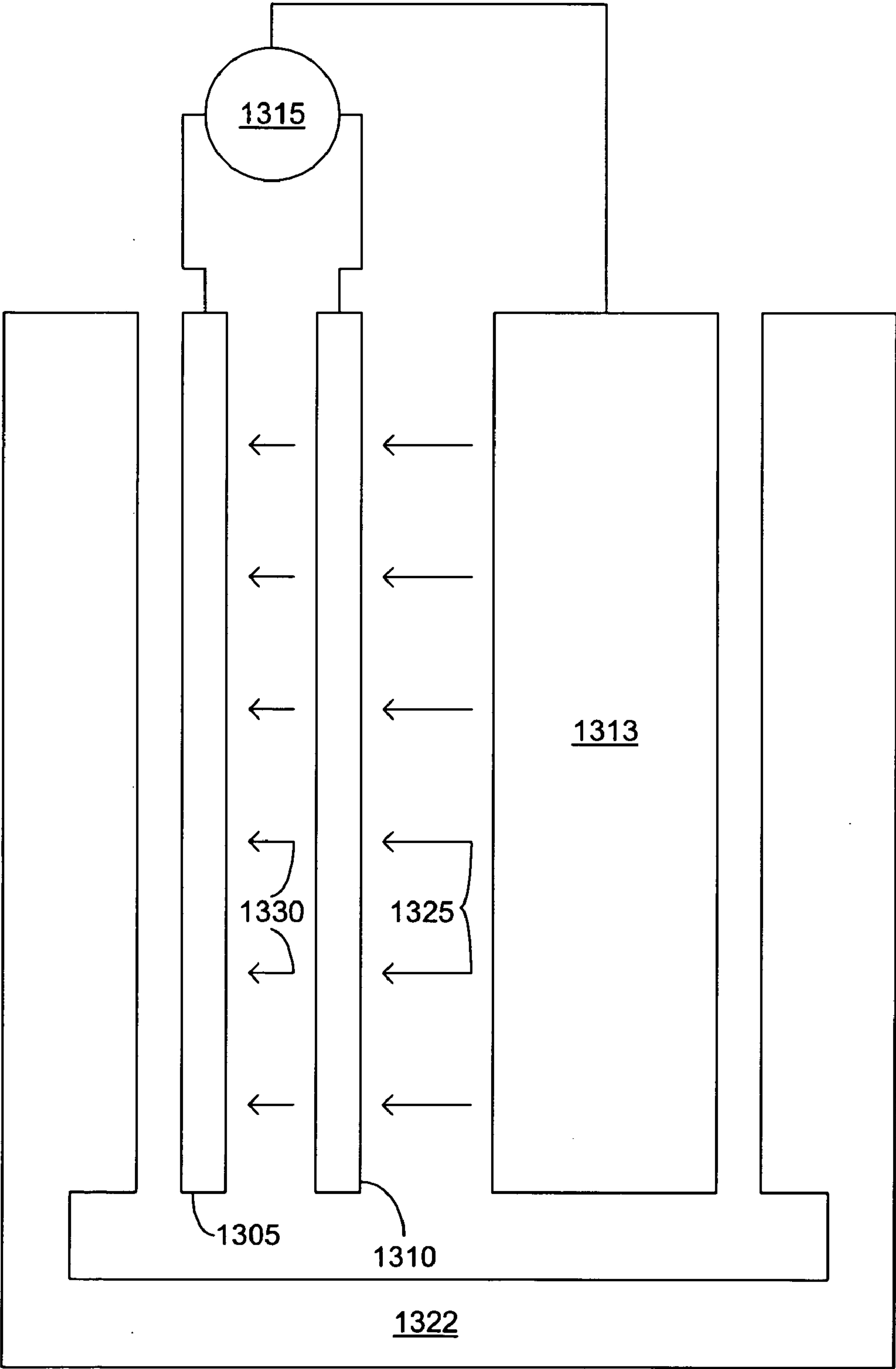


FIG. 13A

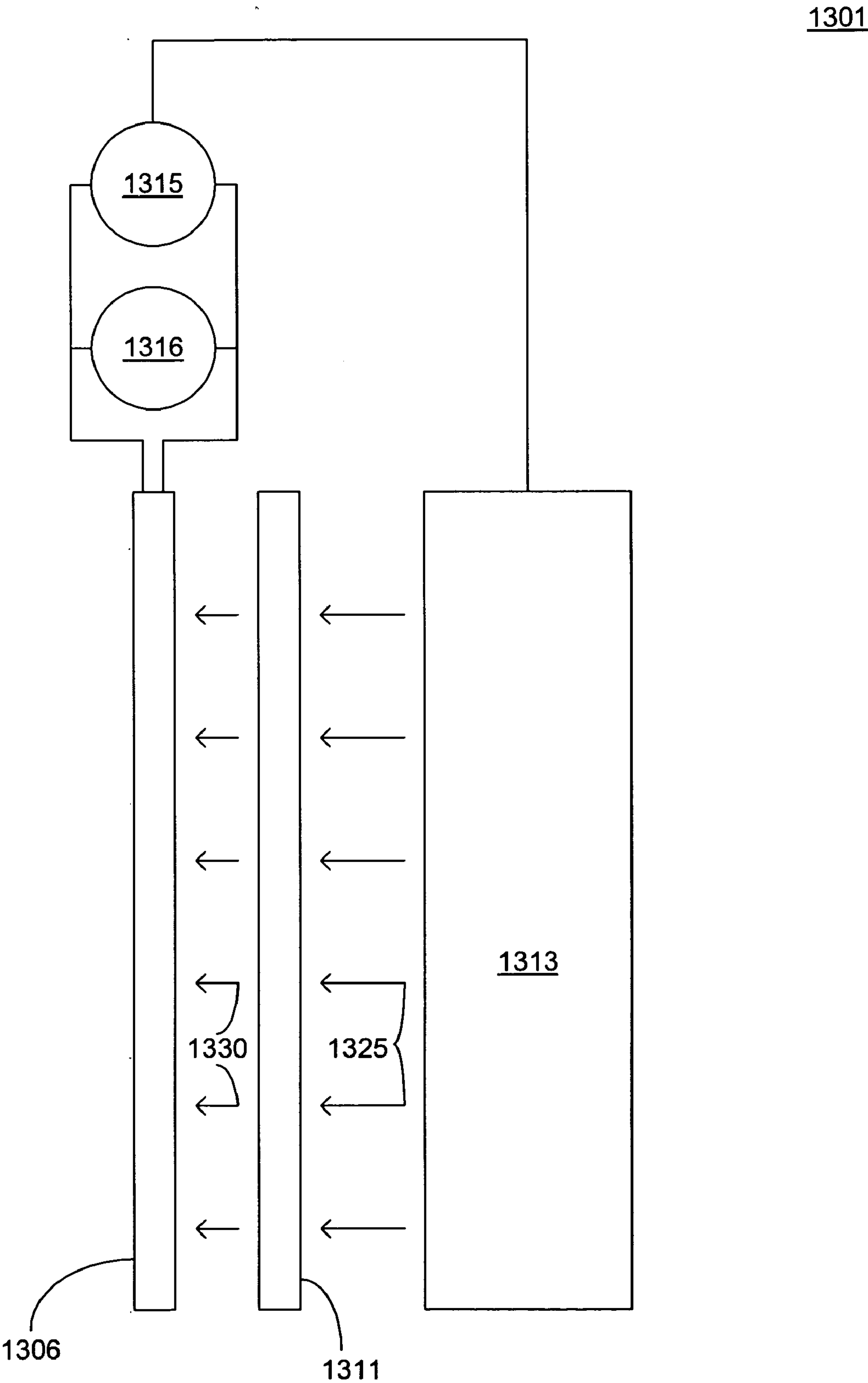


FIG. 13B

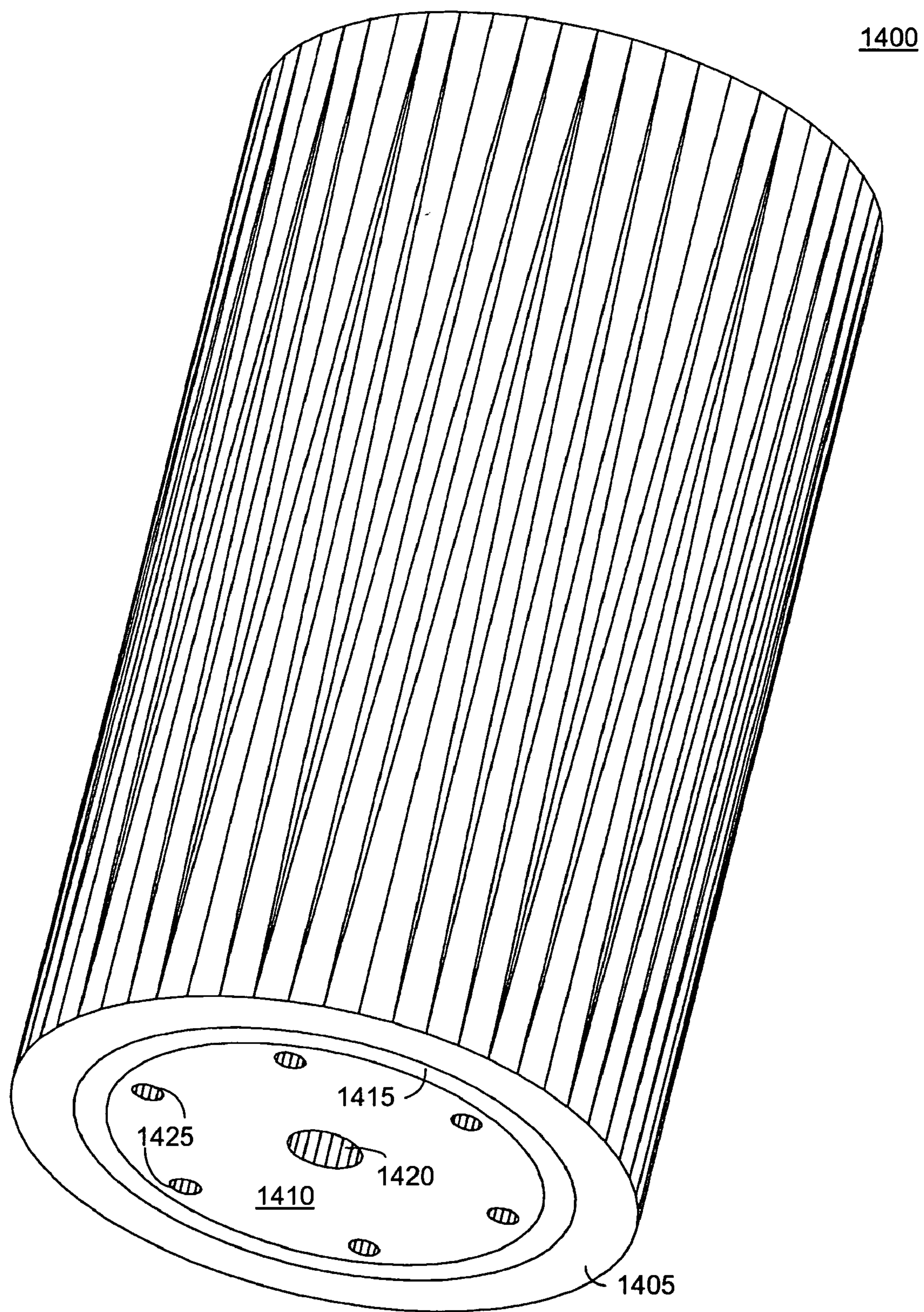


FIG. 14

1500

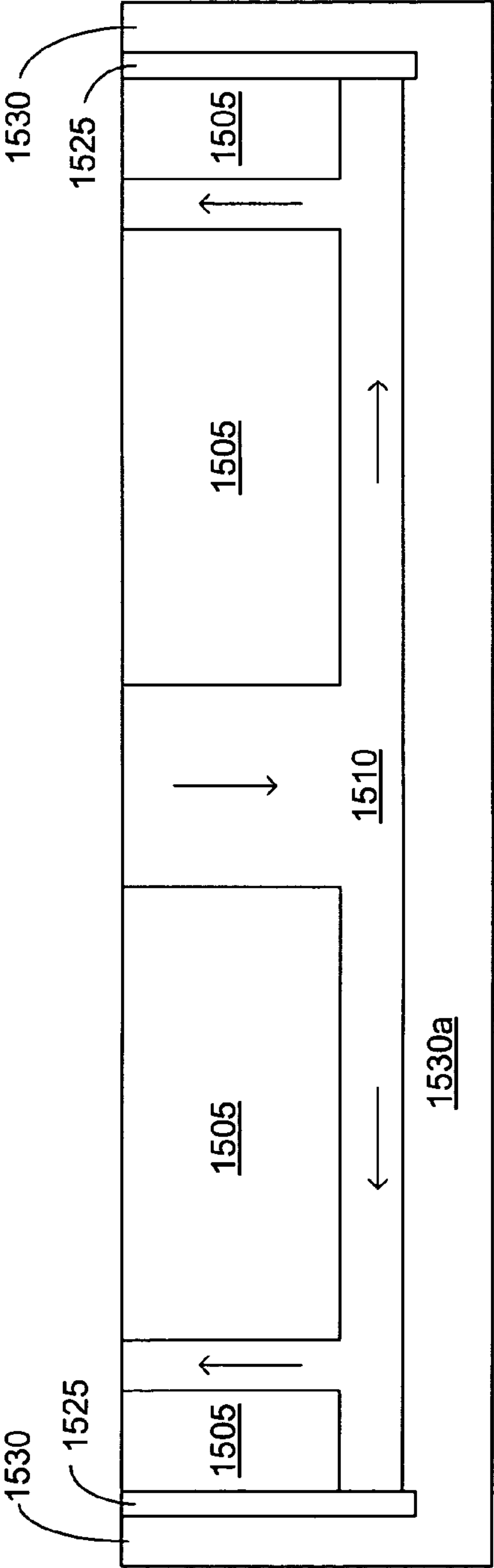


FIG. 15A

1501

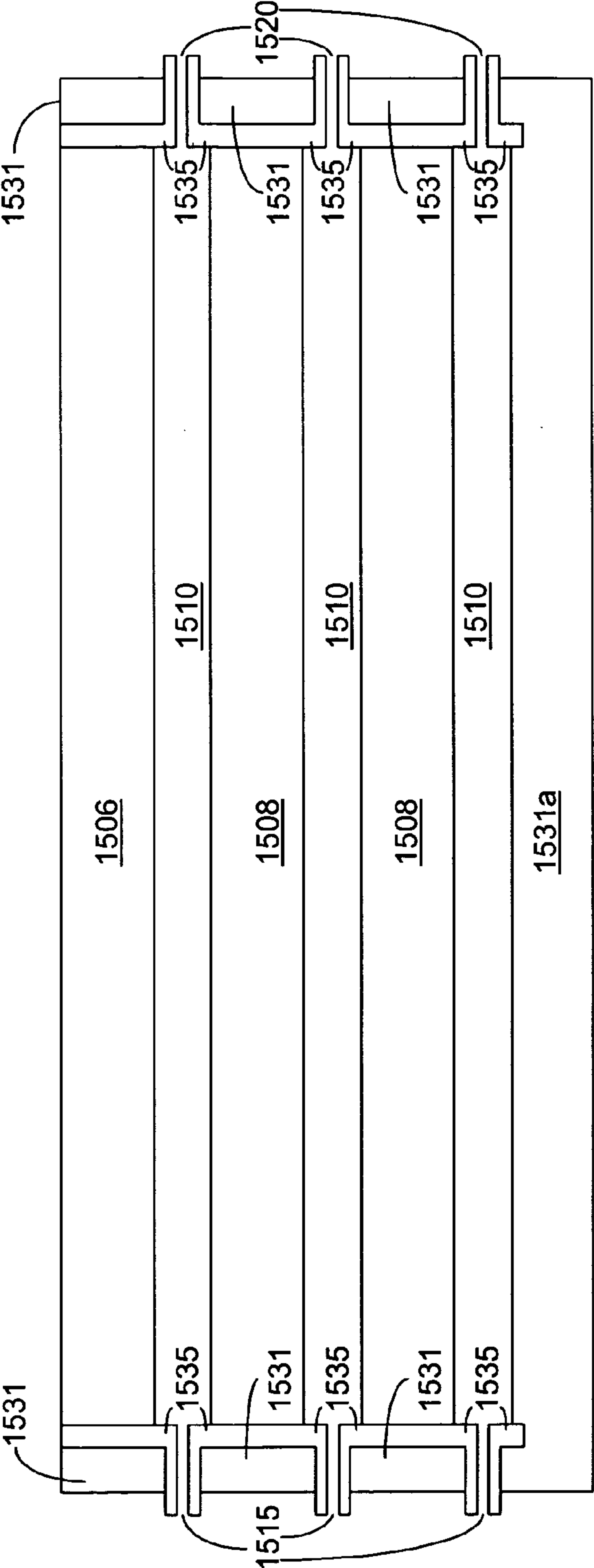


FIG. 15B

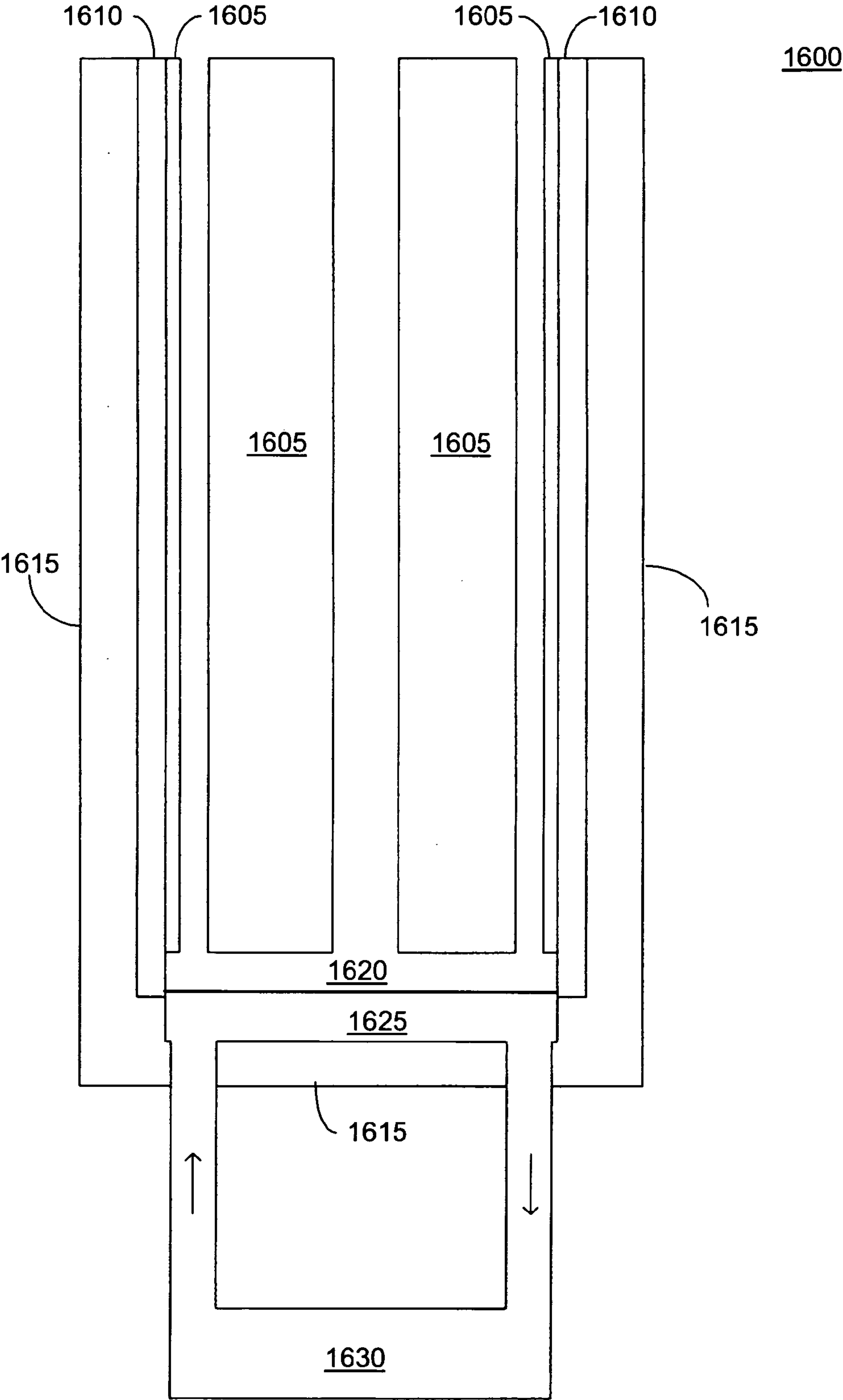


FIG. 16

1700

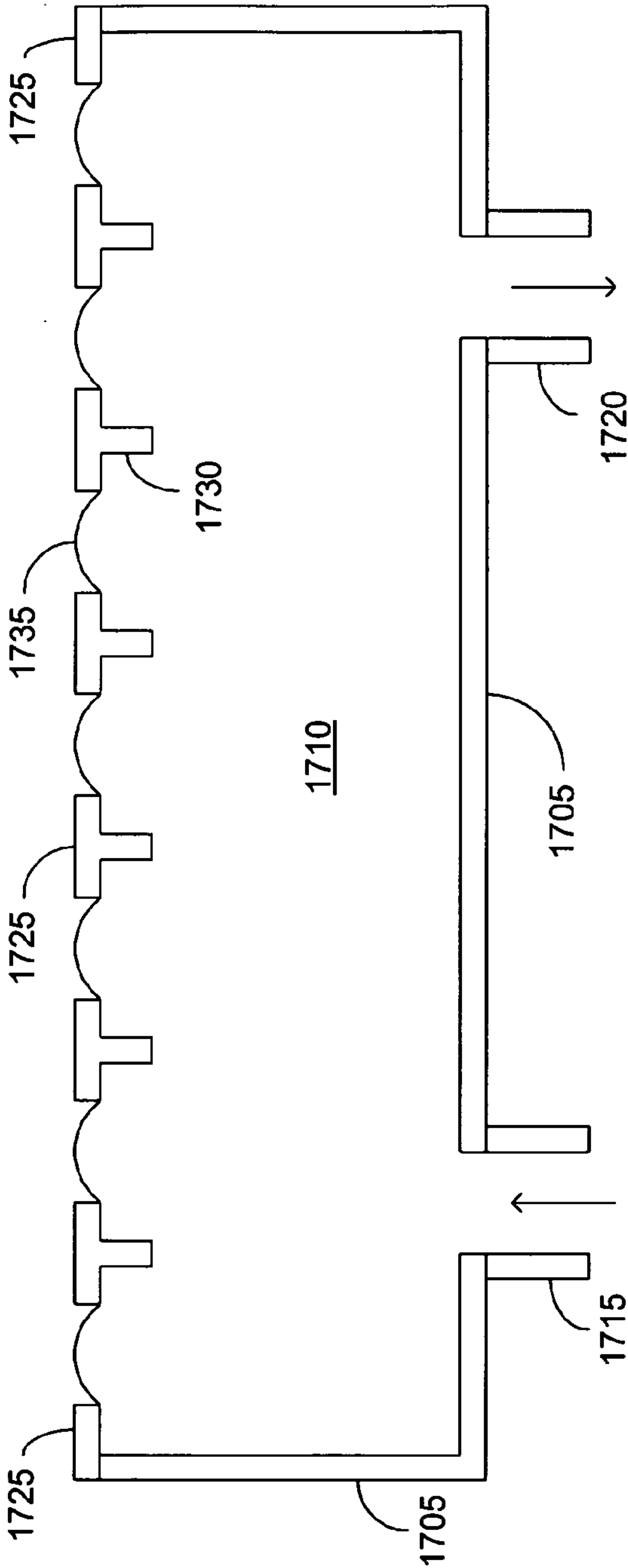


FIG. 17A

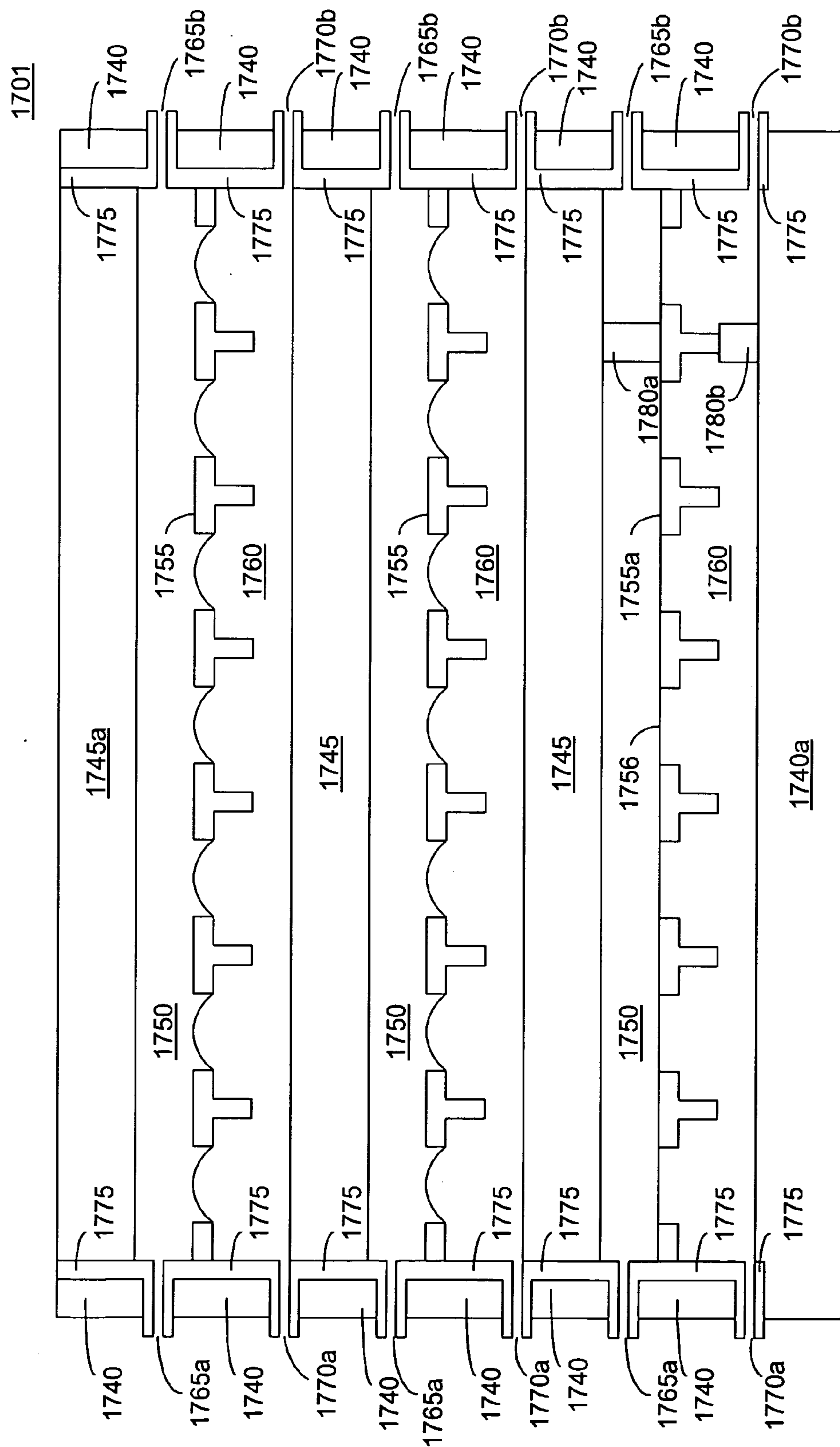


FIG. 17B

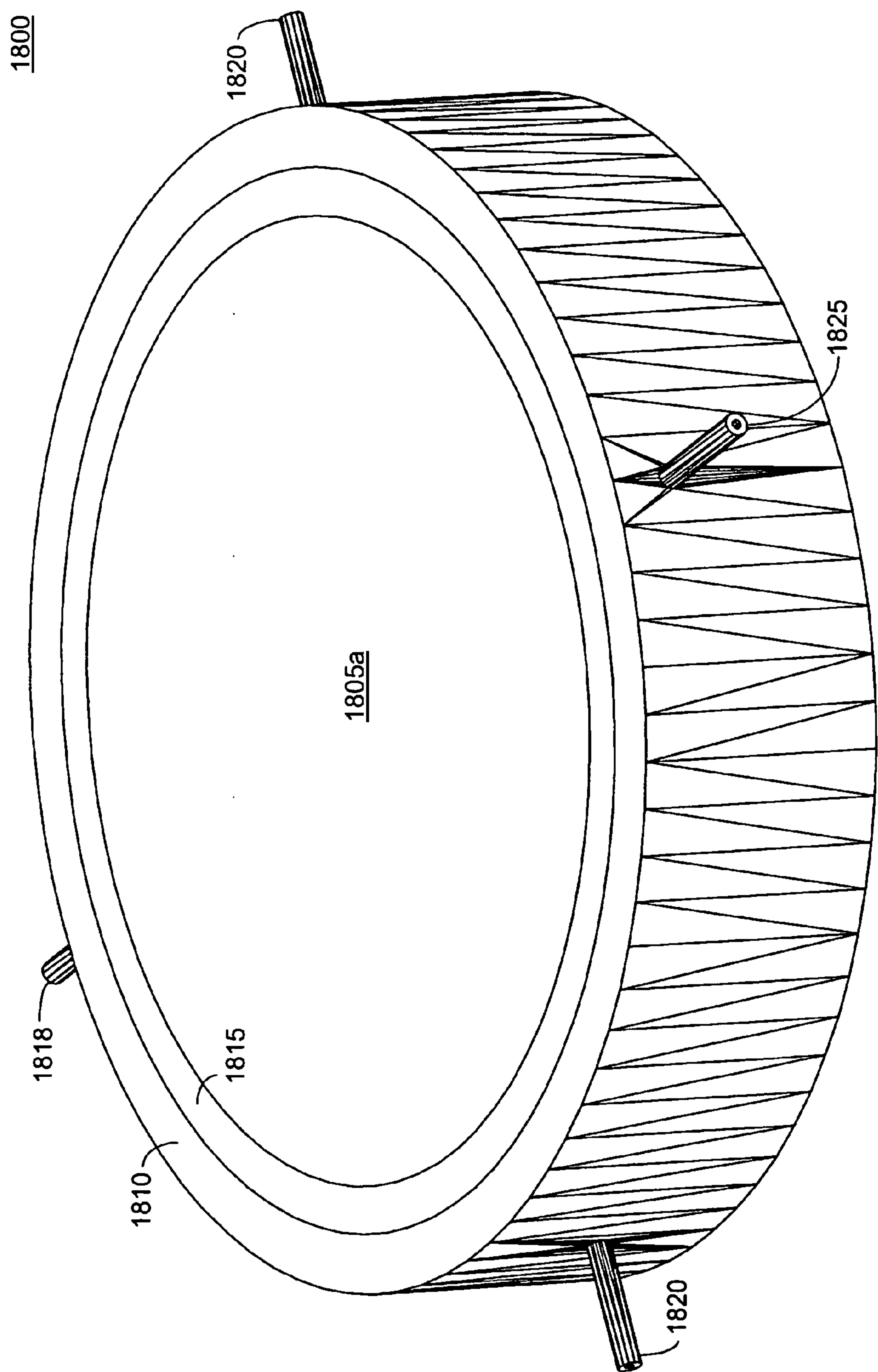


FIG. 18A

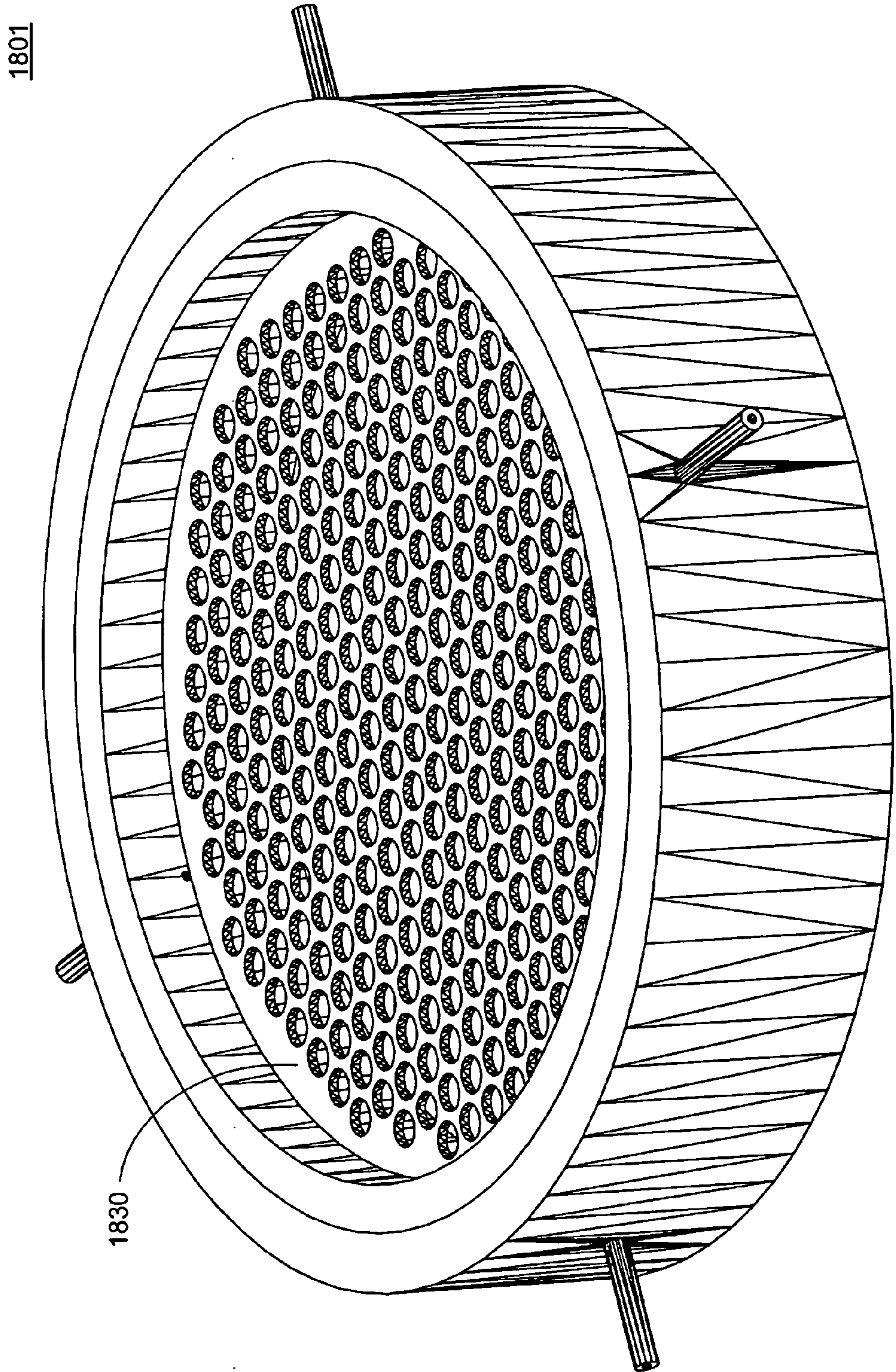


FIG. 18B

1802

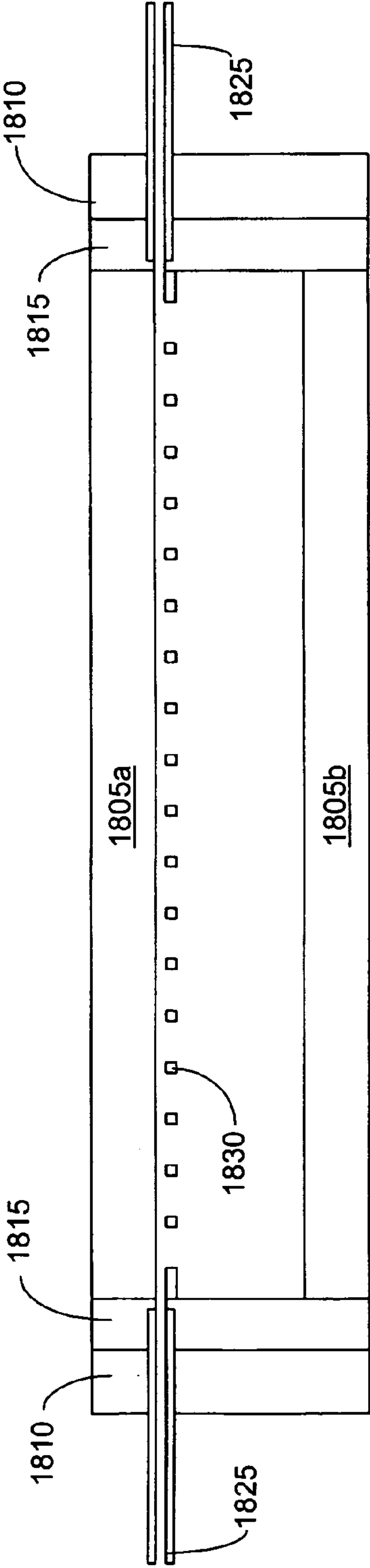


FIG. 18C

1803

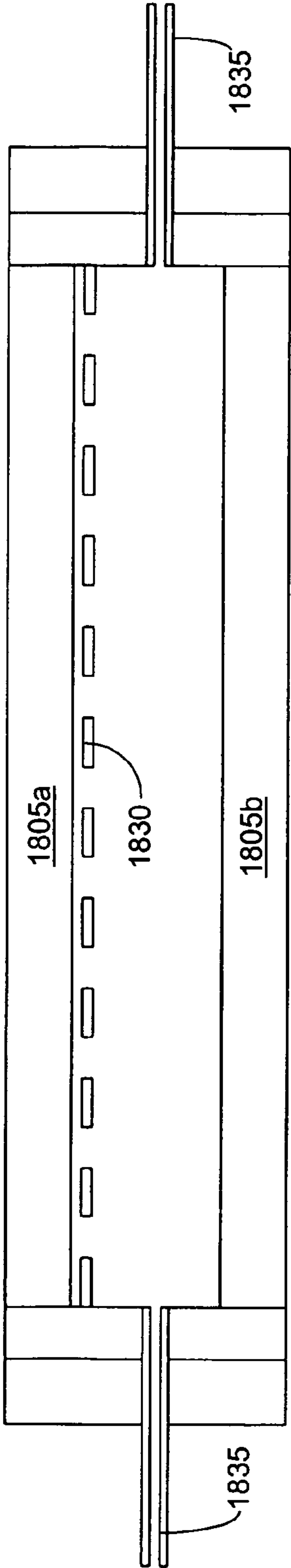


FIG. 18D

1900

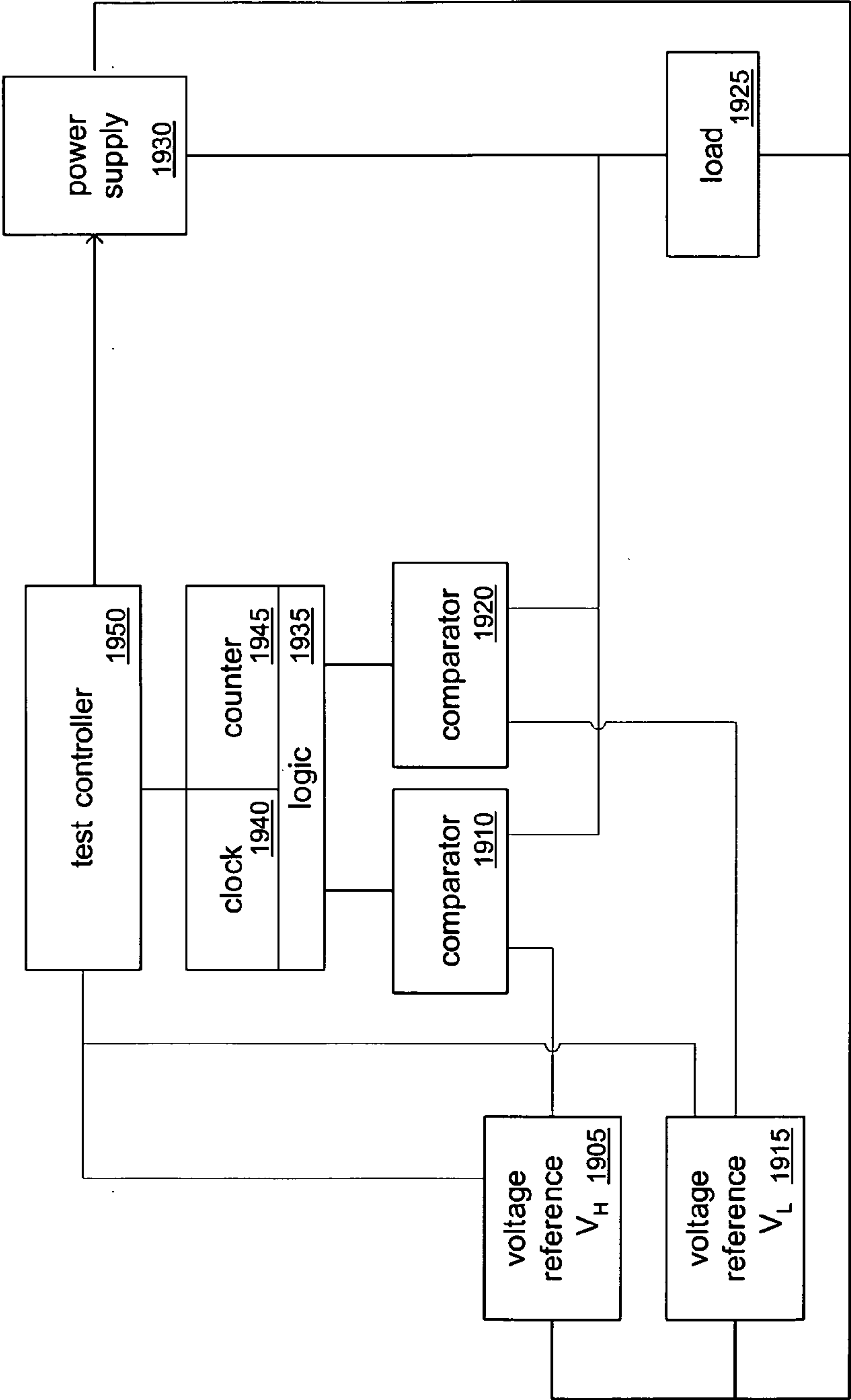


FIG. 19

2000

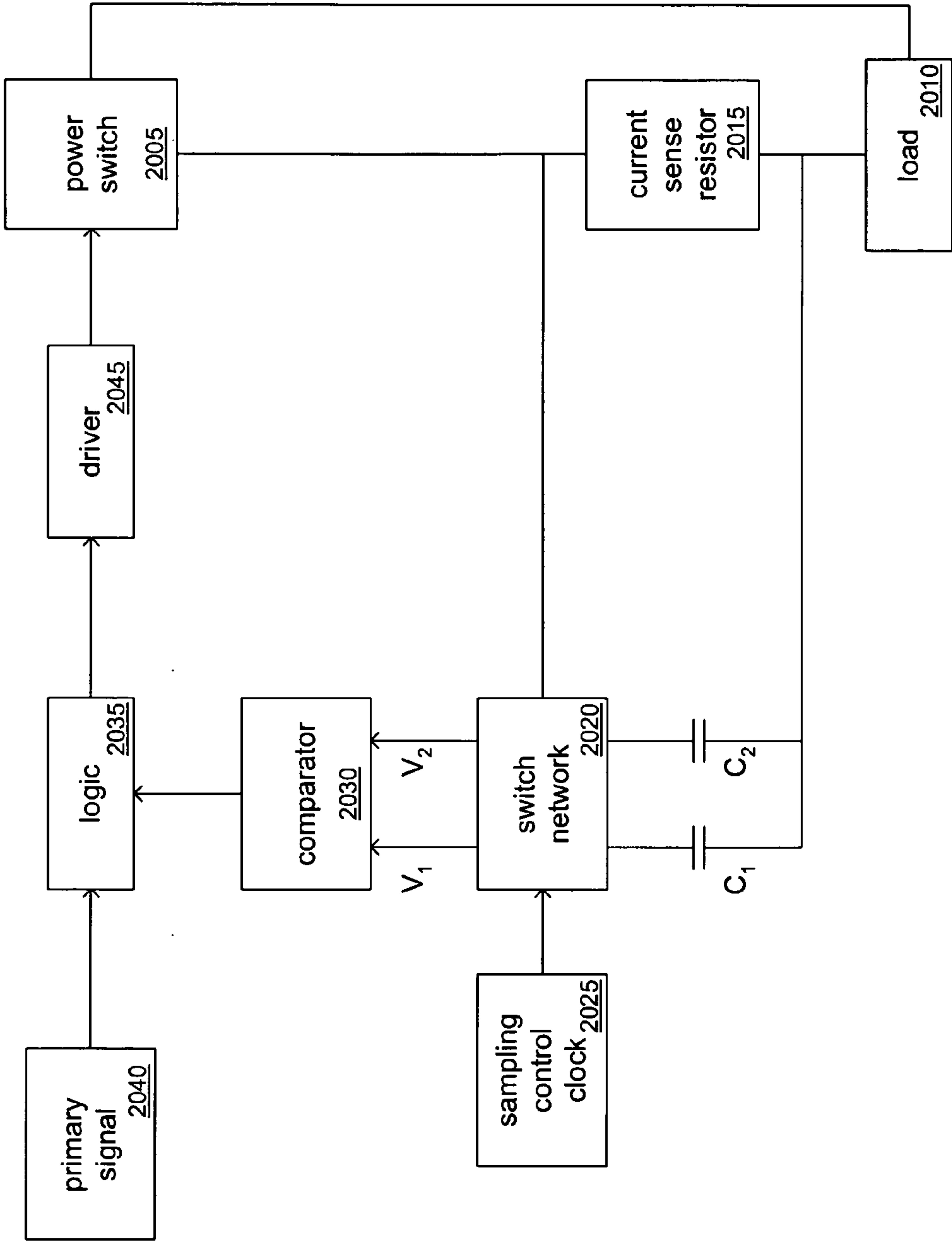


FIG. 20

2100

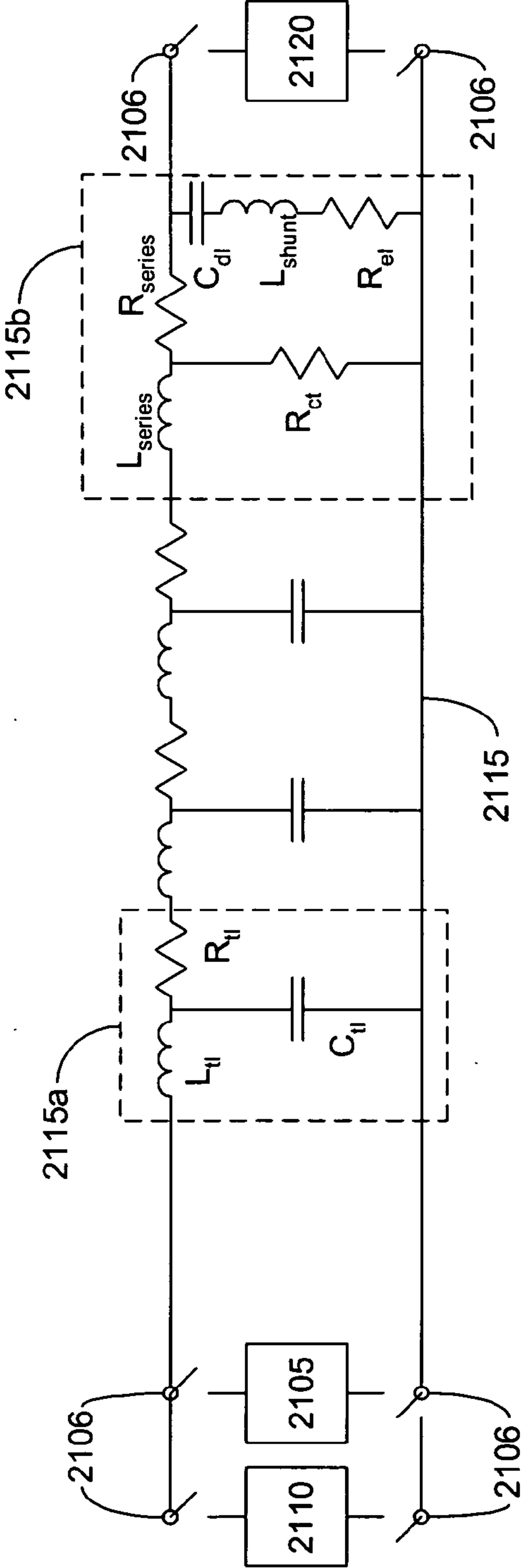


FIG. 21A

2101

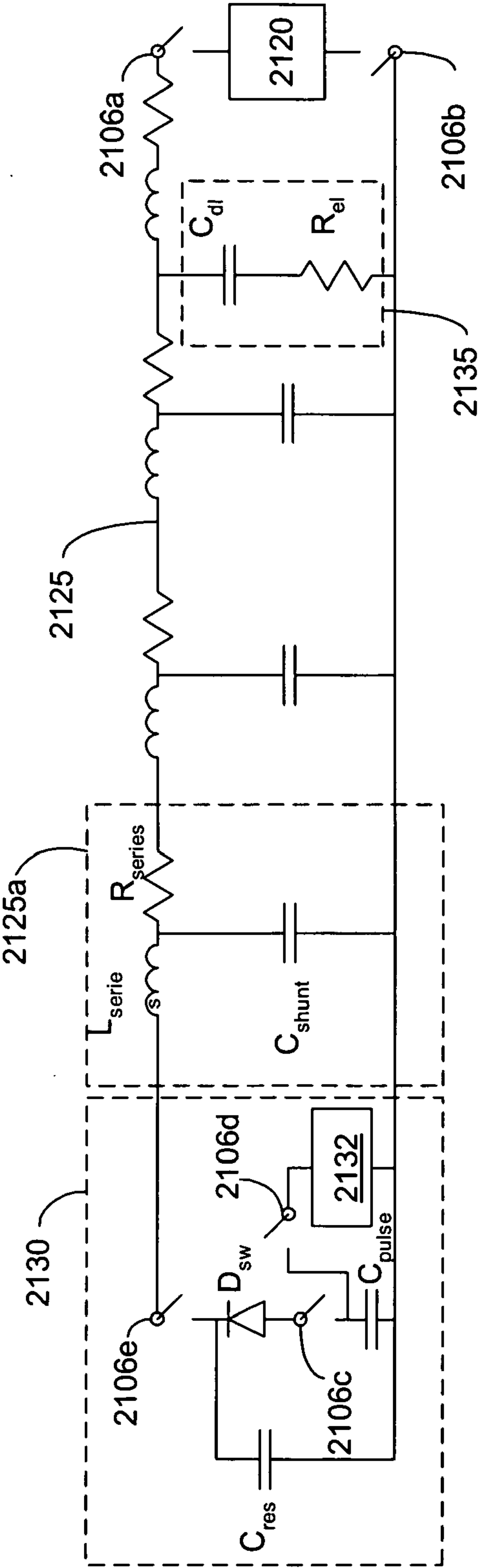


FIG. 21B

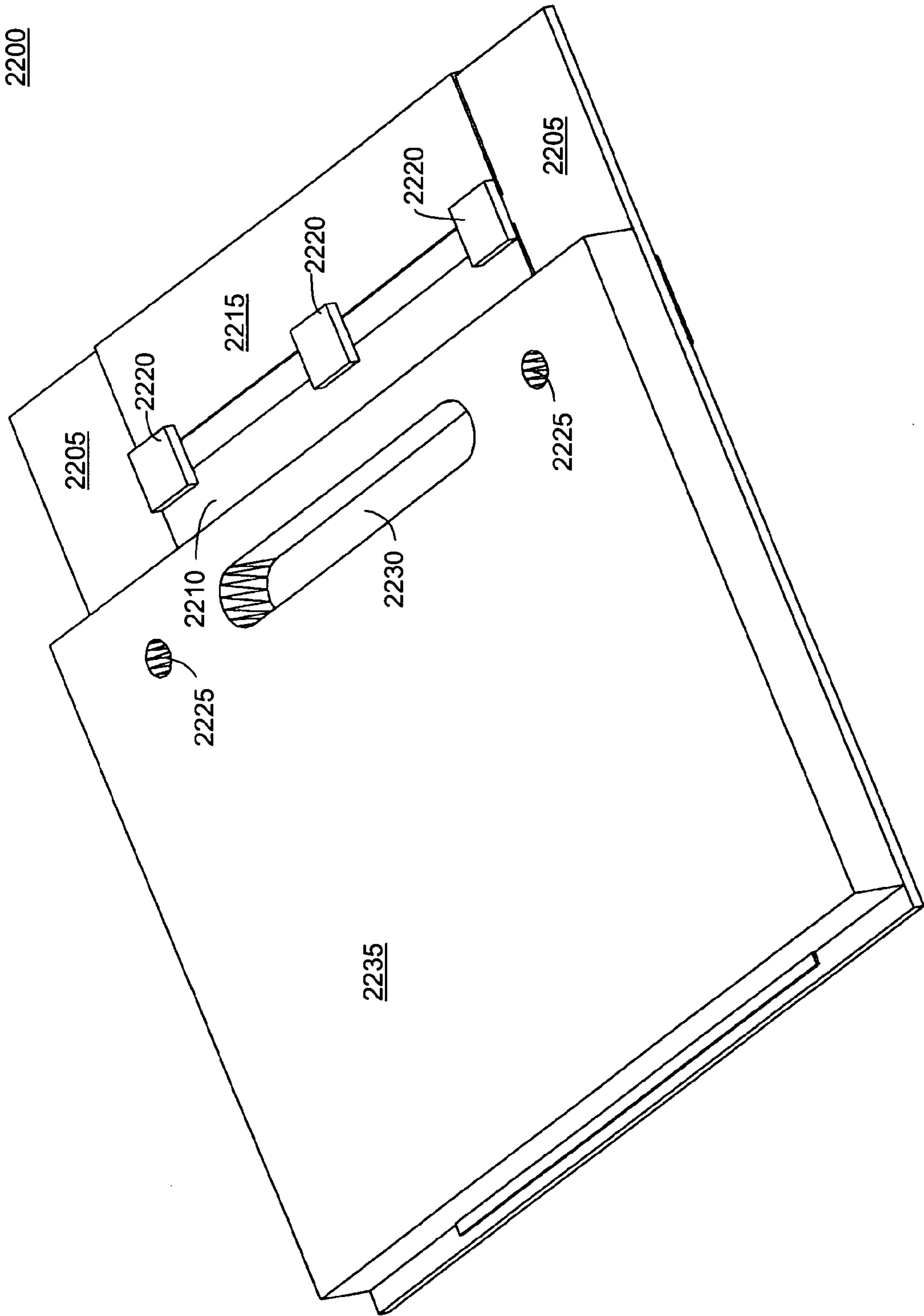


FIG. 22A

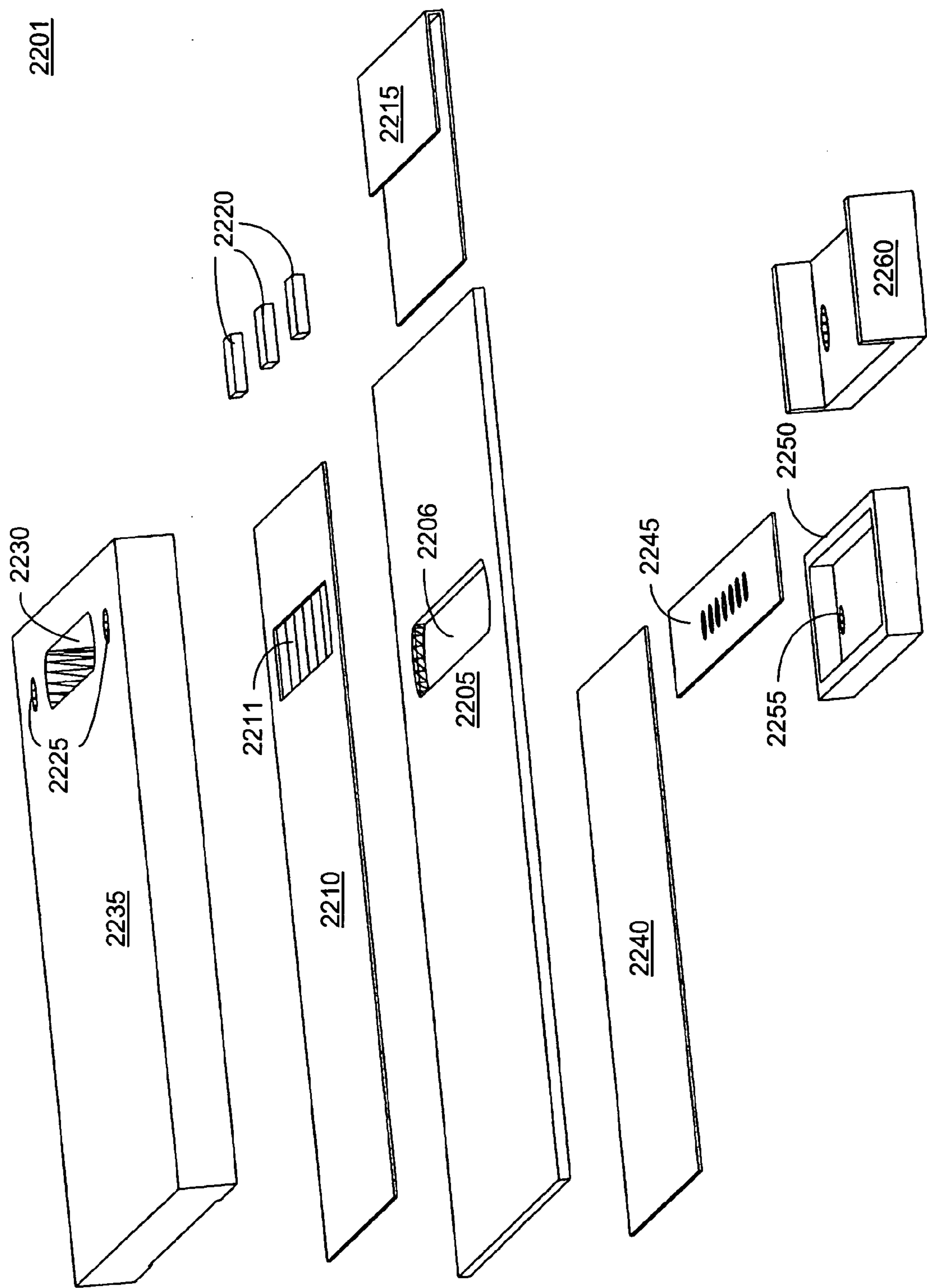


FIG. 22B

2202

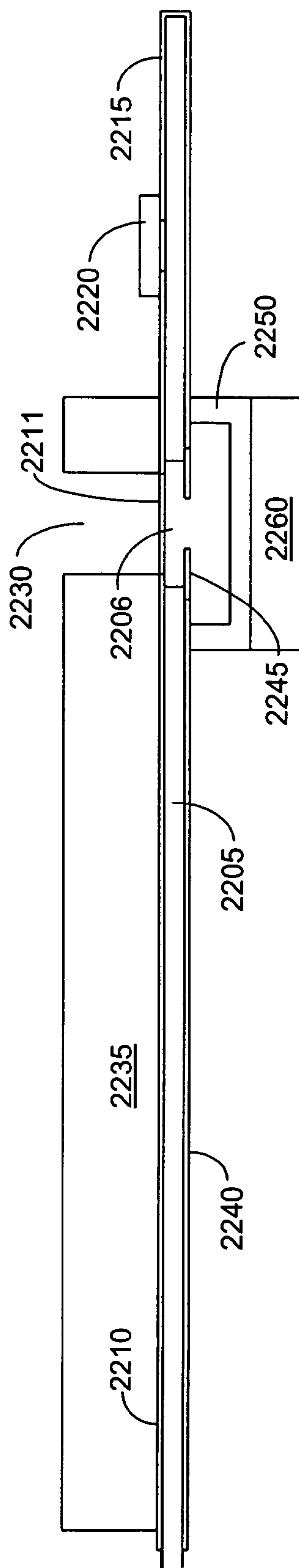


FIG. 22C

2203

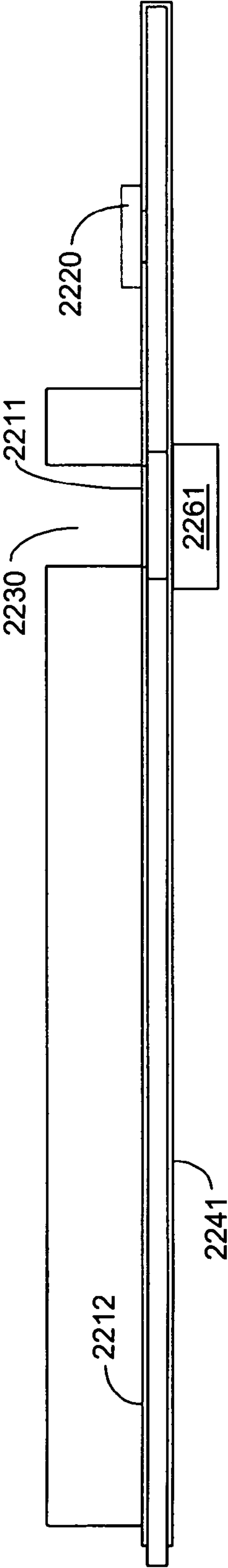


FIG. 22D

2300

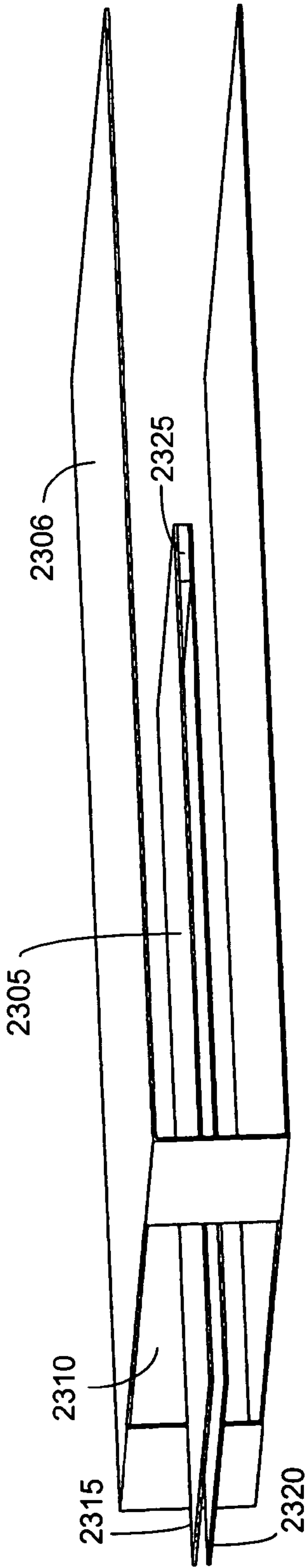


FIG. 23

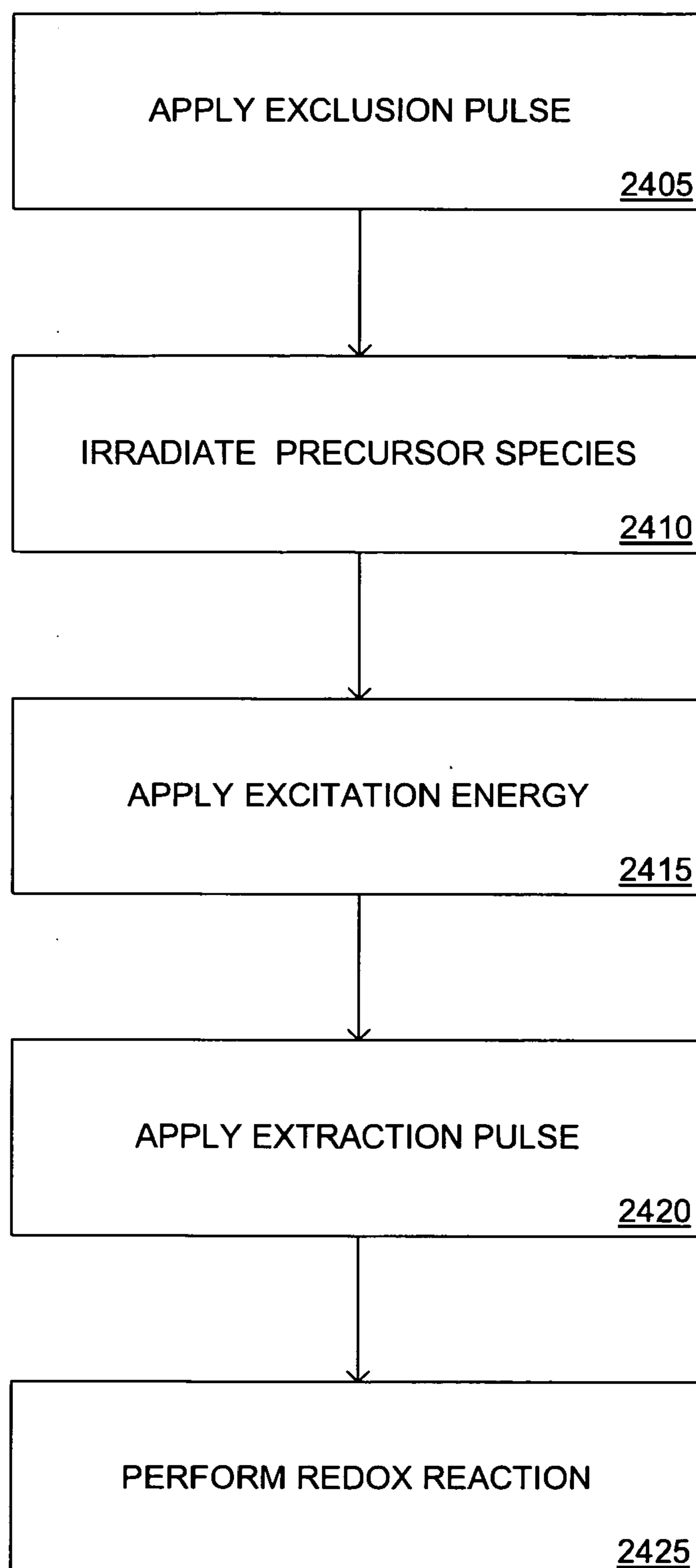
2400

FIG. 24

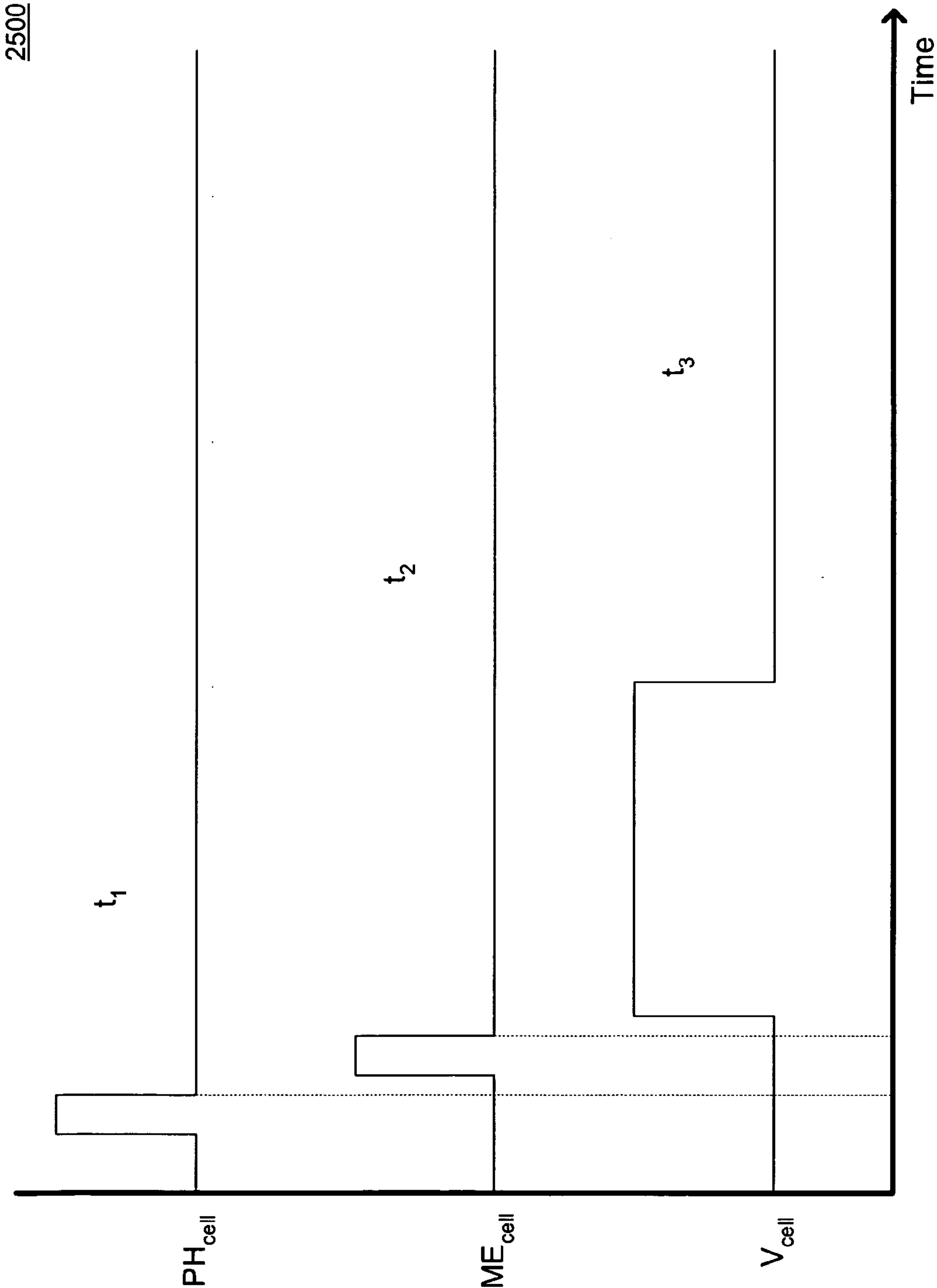


FIG. 25A

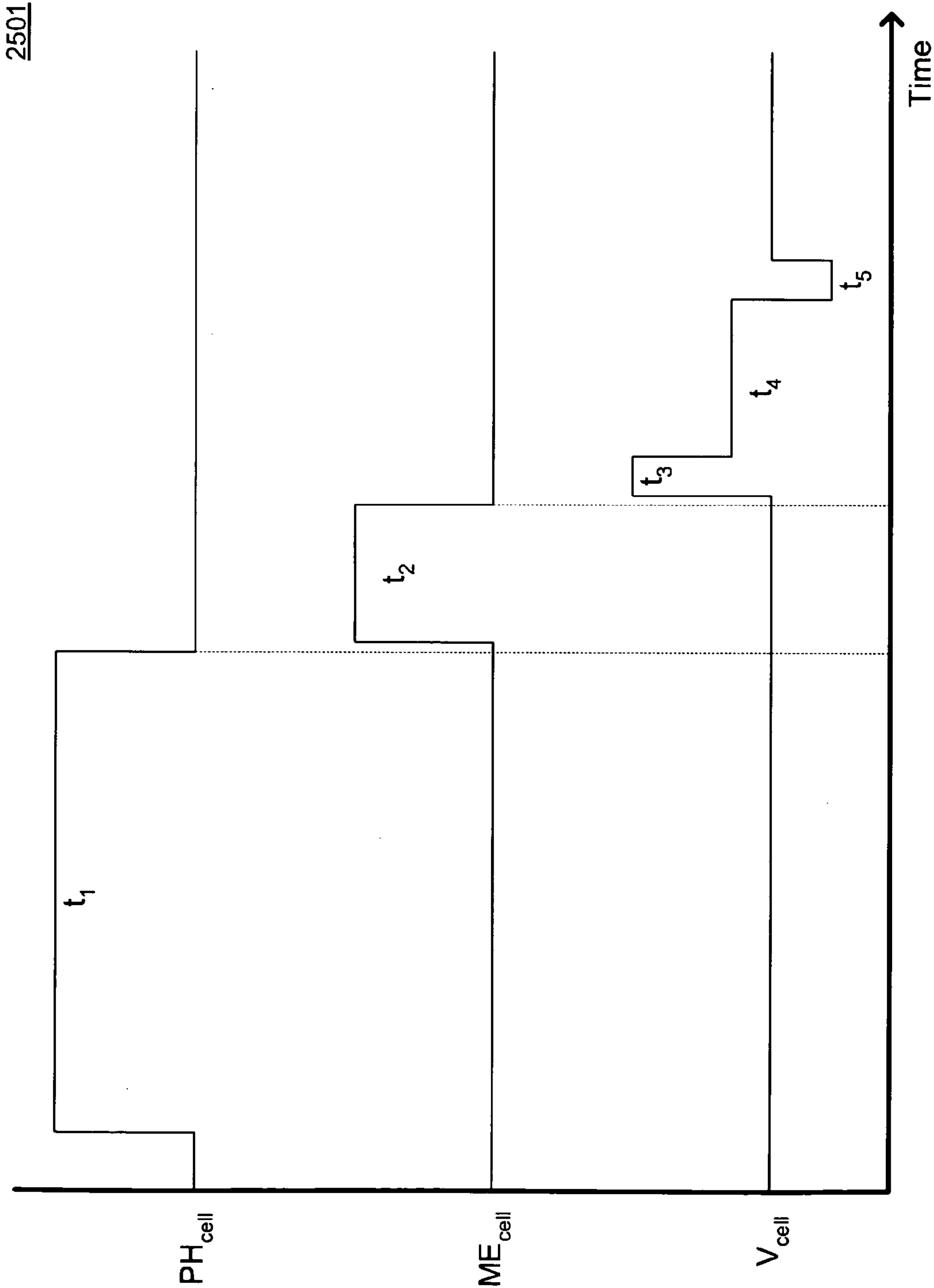


FIG. 25B

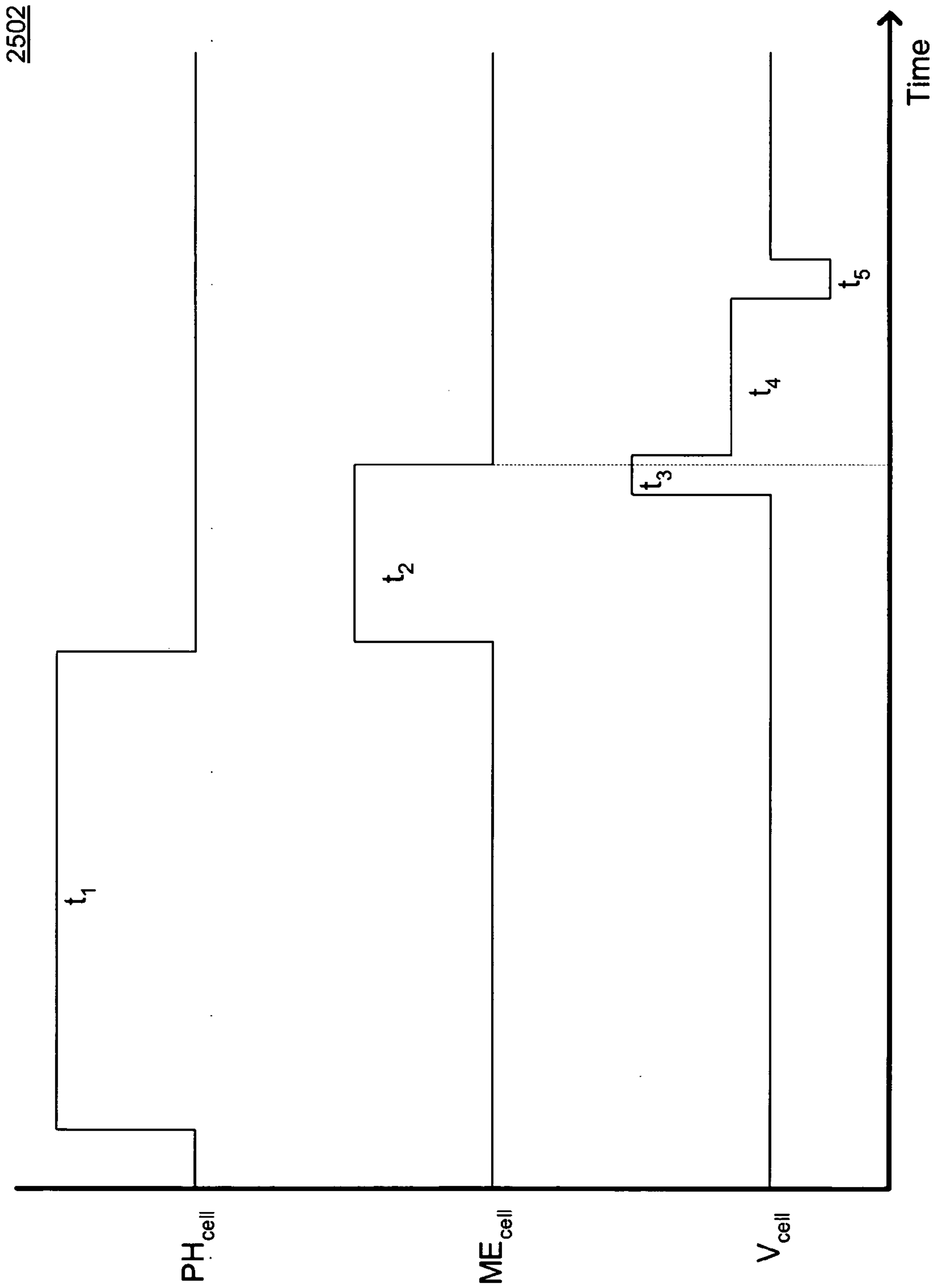


FIG. 25C

2600

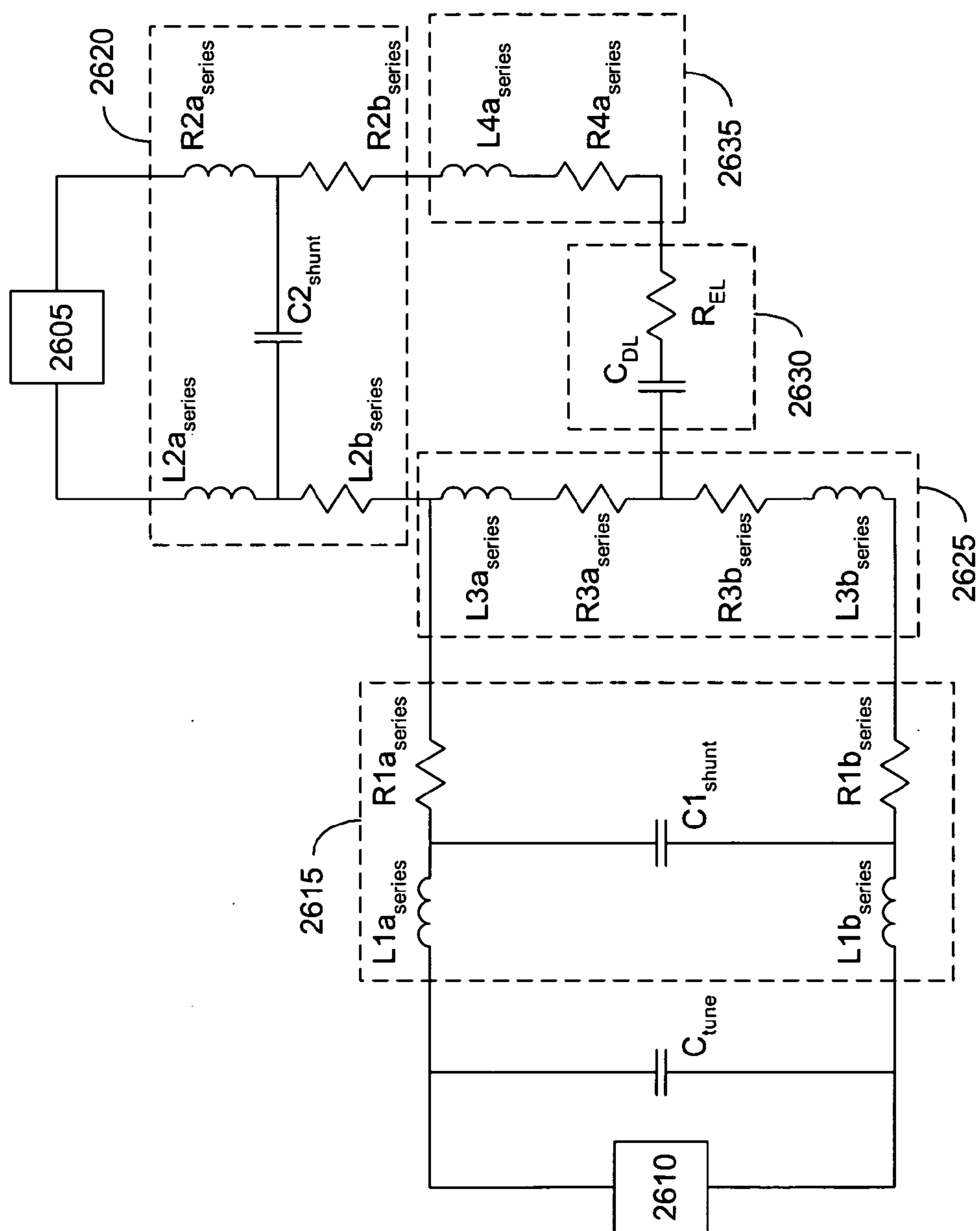


FIG. 26

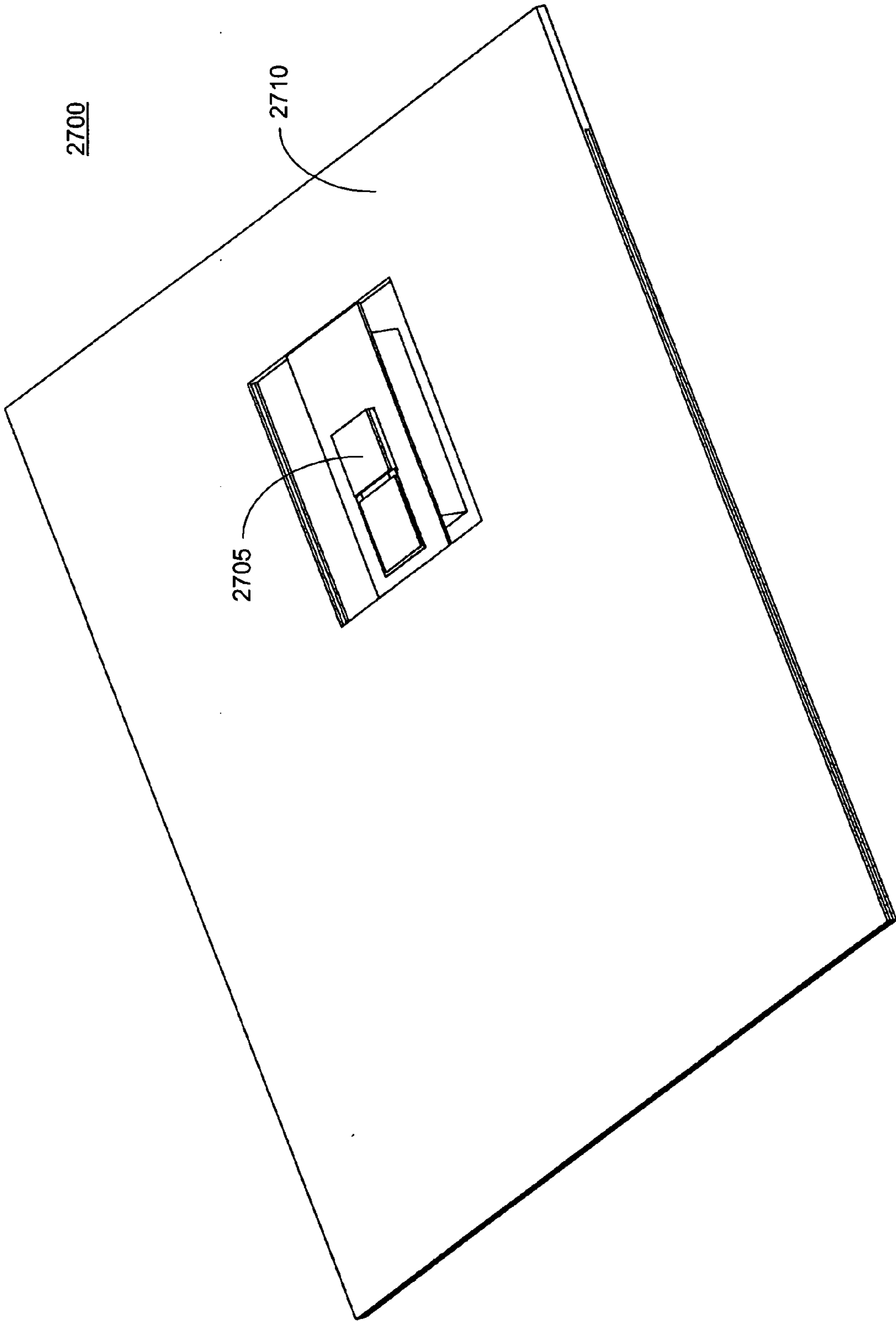


FIG. 27

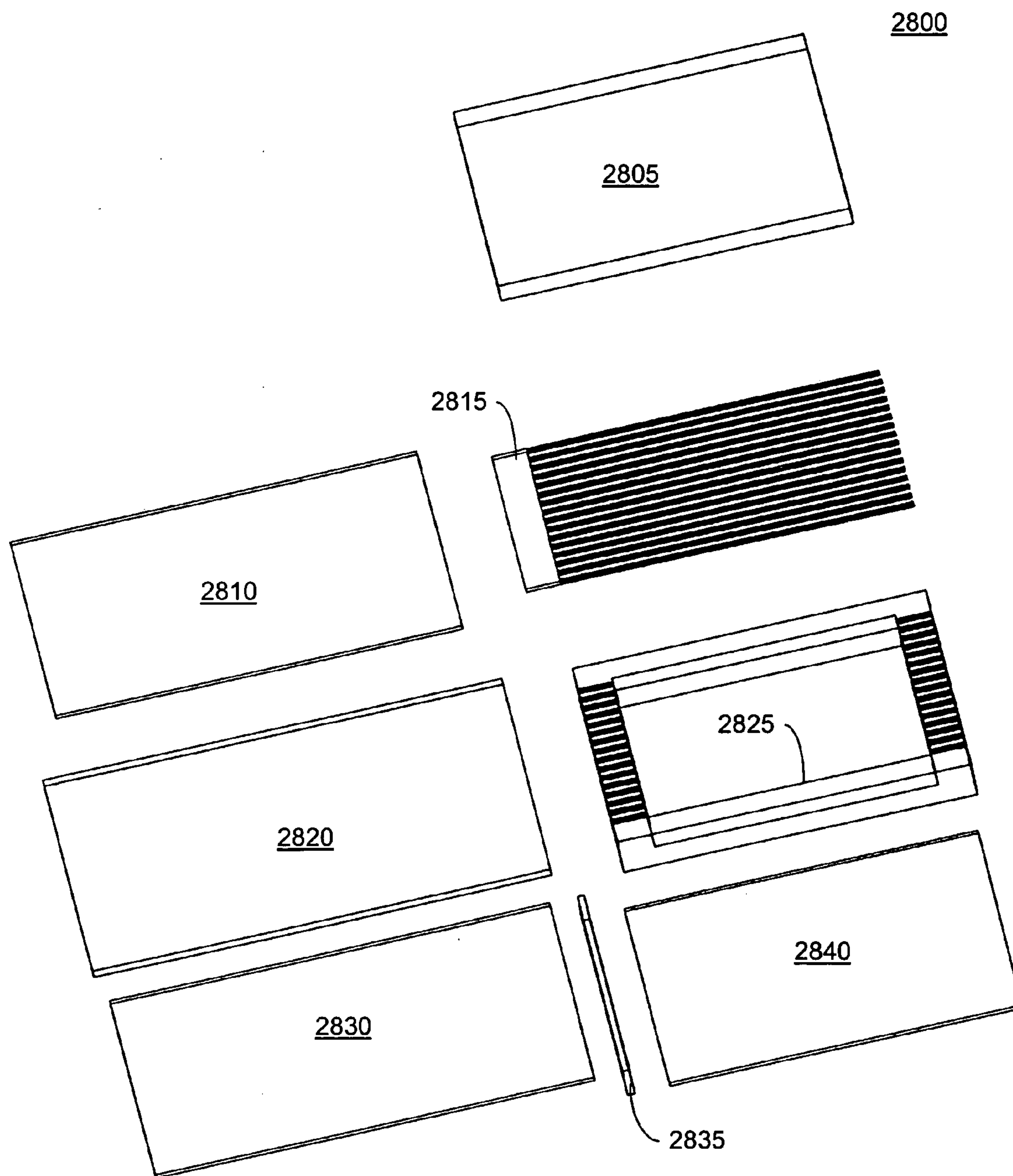


FIG. 28

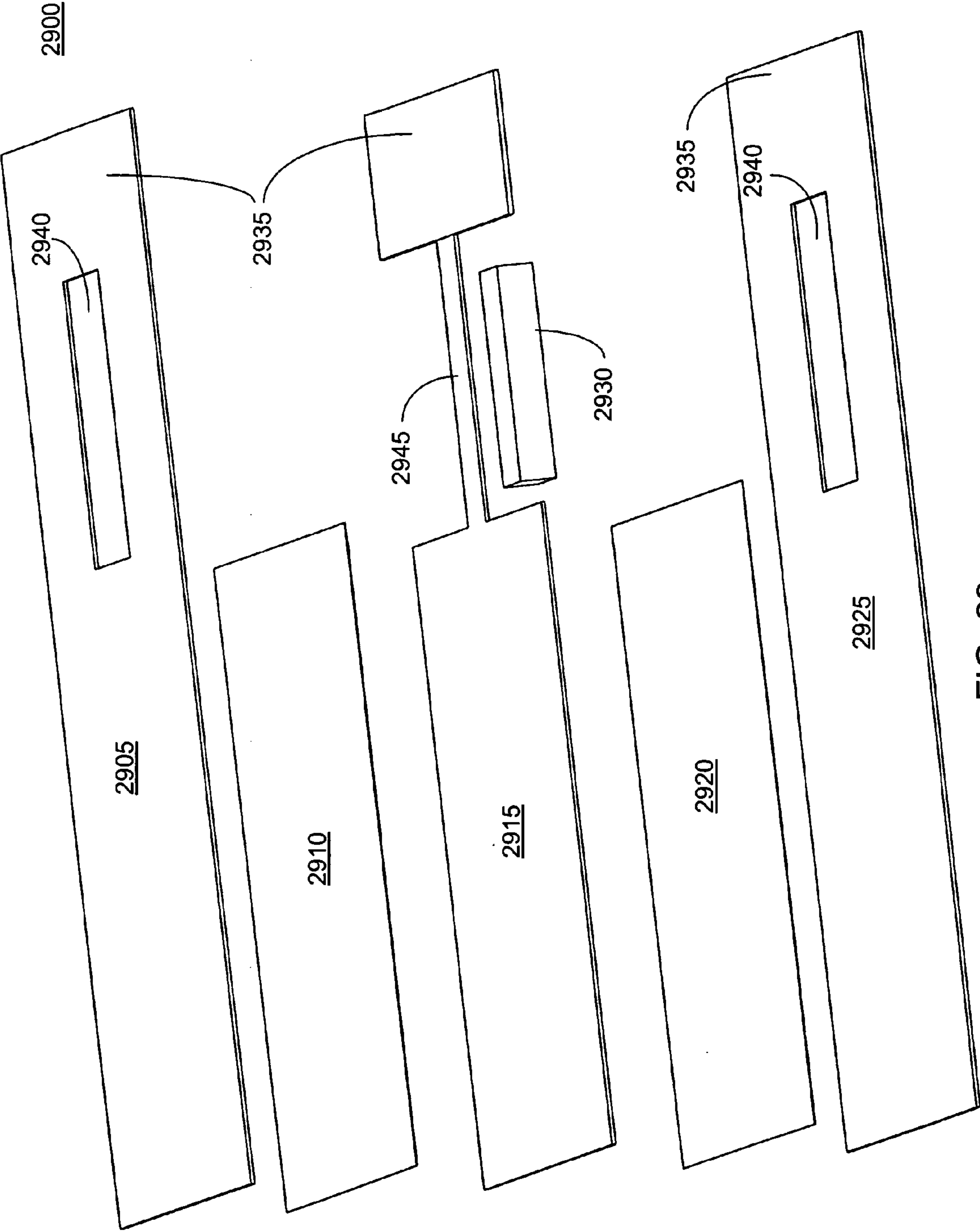


FIG. 29

3000

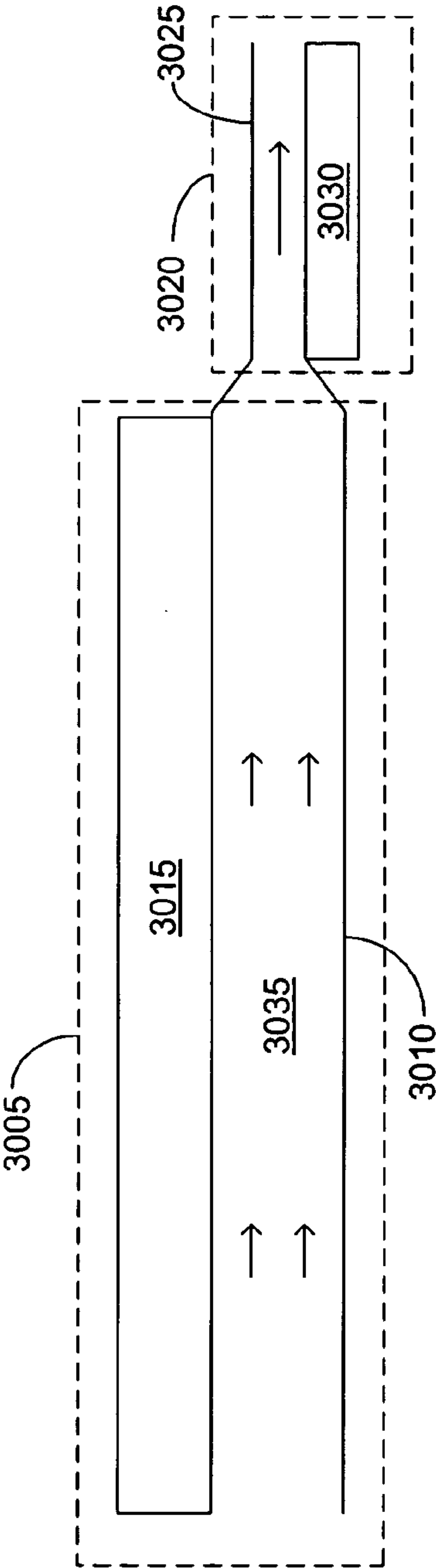


FIG. 30

3100

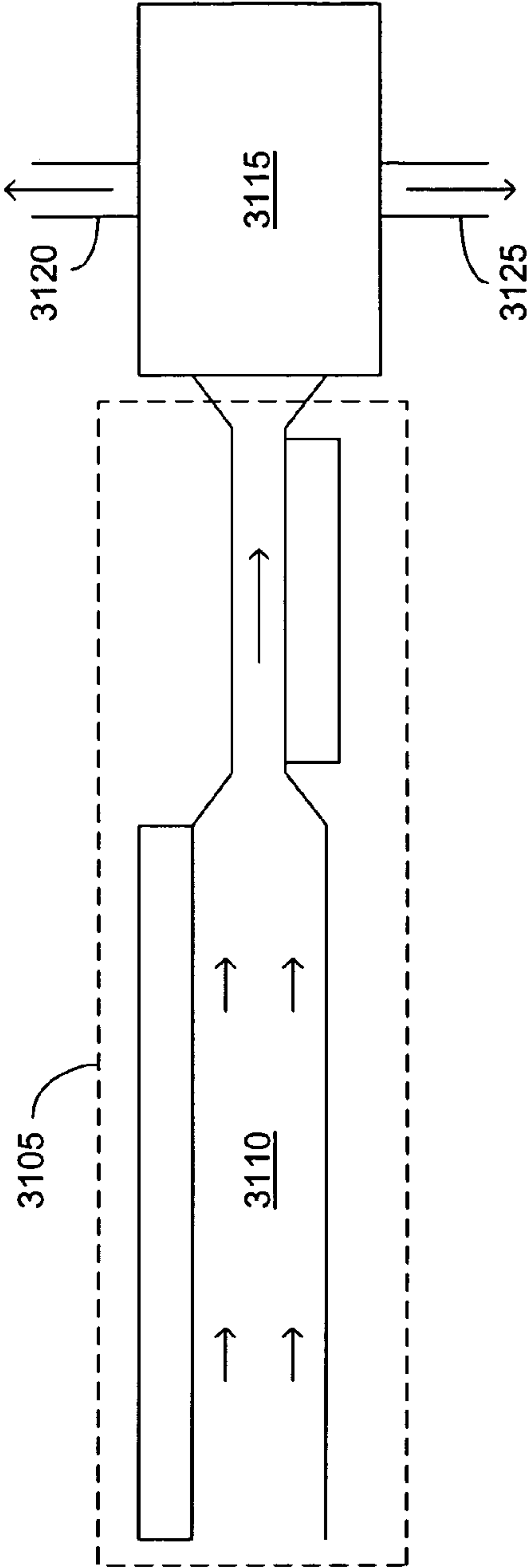


FIG. 31

3200

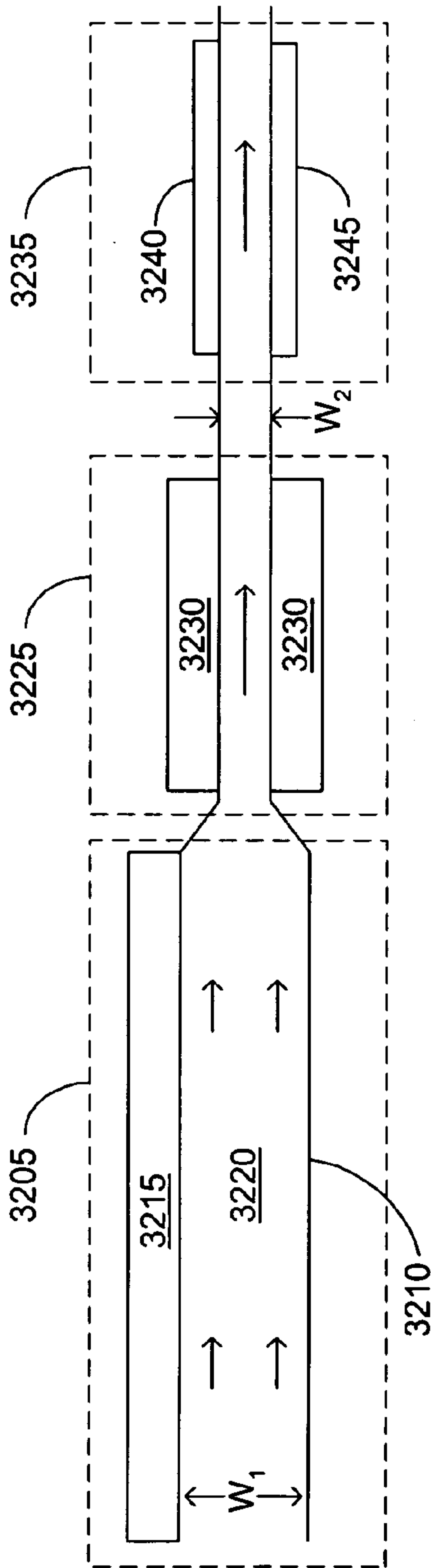


FIG. 32

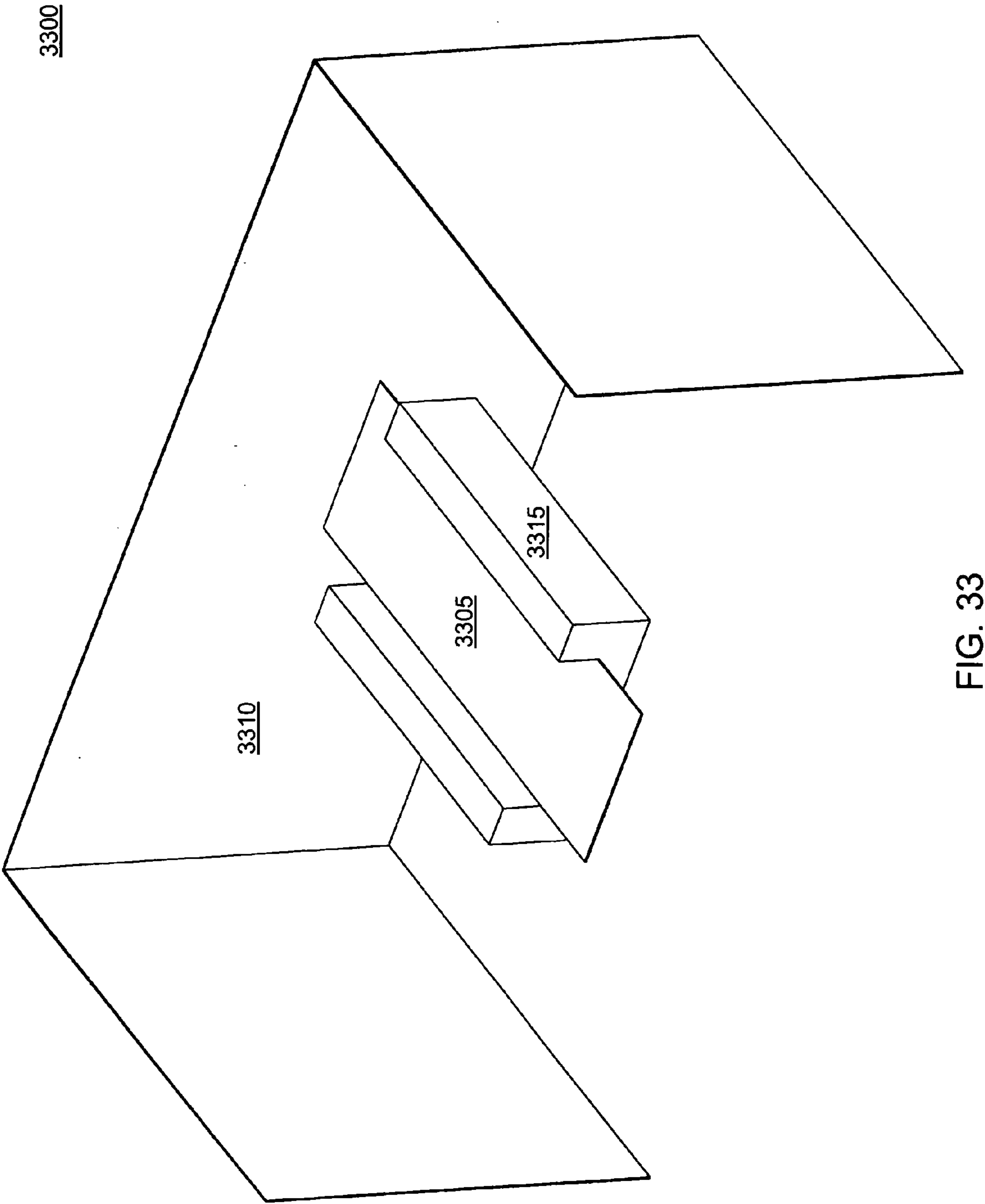


FIG. 33

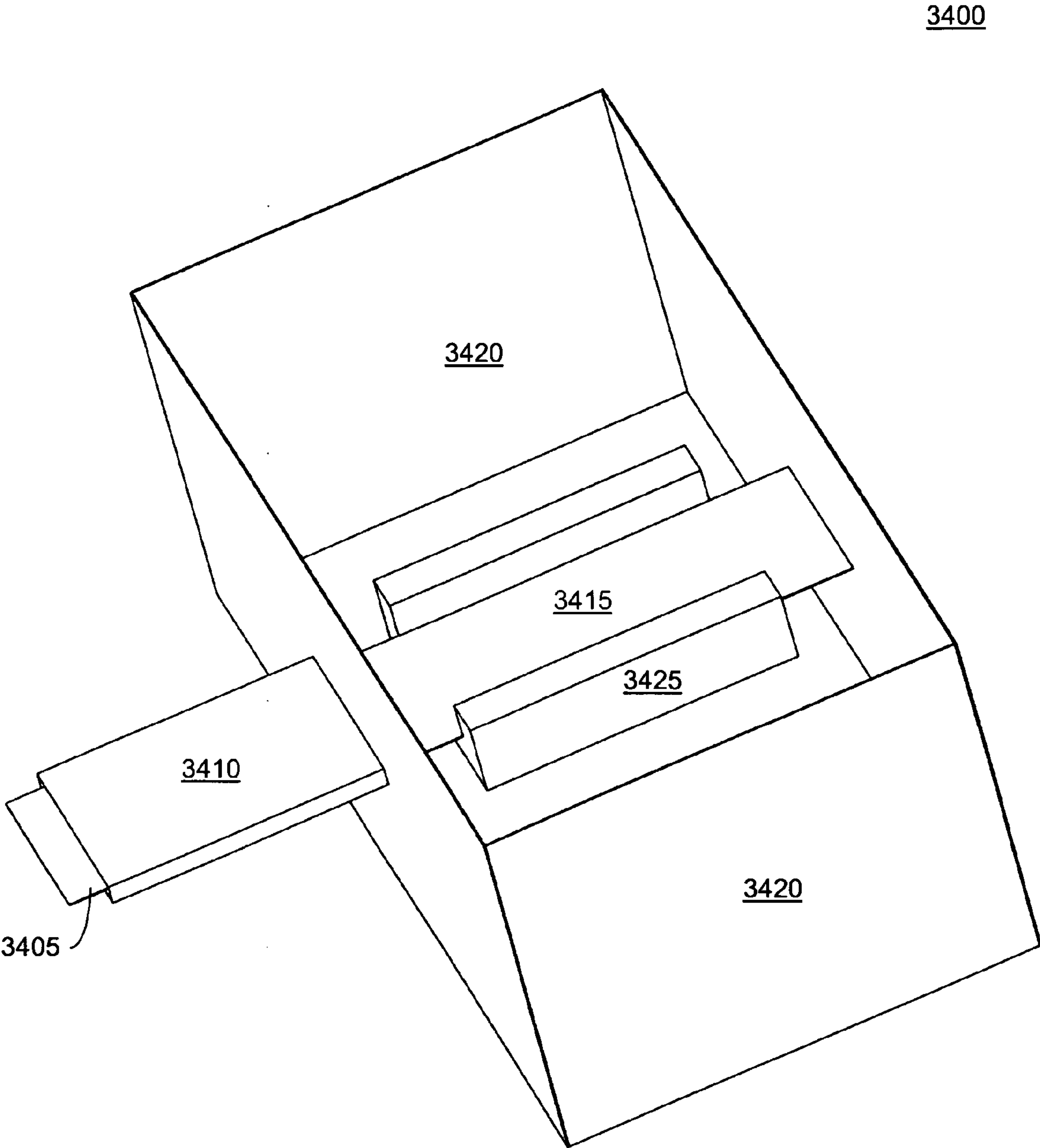


FIG. 34

3500

3535



3530



3505

FIG. 35

SYSTEM AND METHOD FOR ISOTOPE SELECTIVE CHEMICAL REACTIONS

CROSS-REFERENCE TO RELATED APPLICATIONS

[0001] This application claims priority of U.S. Provisional Application No. 60/990,913, filed Nov. 28, 2007.

BACKGROUND OF THE INVENTION

[0002] 1. Field of the Invention

[0003] This invention relates to chemical reactions. In particular, the invention relates to the use of isotope effects to provide selectivity in chemical reactions.

[0004] 2. Description of Related Art

[0005] The transport and reaction of chemical species may be influenced by isotope effects. For example, the classical mass isotope effect (CIE) provides the basis for uranium enrichment by diffusion and by centrifuge. Similarly, isotope effects may be found in electrochemical processes (e.g., the tritium effect), and in photochemical processes (e.g., photoexcitation/photodissociation). The magnetic isotope effect (MIE) may be used to alter the reaction pathways available to chemical reactants, as demonstrated in the photolysis of dibenzyl ketone.

[0006] Although there are a variety of isotope effects that may be harnessed for isotope enrichment and isotope selective reactions, the prior art methods for isotope separation and isotope selective chemical reactions have generally relied upon chemically stable species and focused on the use of a single isotope effect. For example, uranium hexafluoride has been the mainstay for the diffusion and centrifuge uranium enrichment techniques.

[0007] There are many chemical reactant combinations that may be used to provide isotope selective chemical reactions; however, many of these reactions cannot be used in conventional separation processes due to long process timescales. Transport distances on the order of meters and process durations on the order of minutes make it difficult to make use of unstable intermediate complexes or recover unstable products. For example, many photolytic and photoexcitation processes can provide considerable initial differentiation between isotopologues in a gas or liquid, but the differentiation may be quickly lost through environmental interaction (e.g., exchange with neighboring species) or photoemission (e.g., fluorescence).

[0008] Conventional isotope separation processes that are based upon the difference in mass transport of isotopologues (e.g., diffusion and centrifuge) typically have a path length scale on the order of a meter or more, and thus have a relatively large minimum size for a functional unit. For many applications it would be desirable to have a small low-cost functional unit that could be used in a scalable array.

[0009] The differences in spin behavior between magnetic and nonmagnetic nuclei provides a basis for isotope separation and selective chemical reactions. For example, the behavior of a radical pair may be controlled by selective spin conversion and spin locking (e.g., manipulation of the Zeeman and hyperfine interactions by applied DC and RF magnetic fields). Microwave pumping has typically been done using microwave tubes as sources and beaming the microwave energy into the working material through waveguides. The combination of a tube and waveguide has the disadvantage of low efficiency, particularly if the dimensions of a

working cell are below the cutoff frequency of the waveguide. The prior art of spin conversion does not provide efficient means for producing RF magnetic fields in highly localized regions.

[0010] Thus, there is a need for an isotope selective chemical reactor that uses a combination of isotope effects. There is also a need for a small chemical reactor that can take advantage of short-lived intermediates and products. It is also desirable that a small chemical reactor serve as the basic unit for a scalable array.

BRIEF SUMMARY OF THE INVENTION

[0011] Accordingly, a system for isotope selective chemical reactions is described herein. Two or more isotope effects are combined within a reaction cell to produce transient differentiation of isotopologues through excitation or chemical reaction followed by rapid separation or conversion.

[0012] In an embodiment of the present invention, a current conductor adjacent to a fluid carries an excitation current produced by a switching source. The excitation current produces an alternating magnetic field adjacent to the conductor that interacts with a population of isotopologues within the adjacent layer to produce a population of reaction intermediates or products that have an isotopic composition different from the initial isotopologues.

[0013] In a further embodiment, an electrolytic power supply is also connected to the conductor and provides a voltage that may be used to selectively attract and electrolyze a portion of the reaction intermediates or products within an adjacent fluid electrolyte layer.

[0014] In another embodiment, the adjacent layer is subjected to electromagnetic radiation to provide excitation or dissociation. The electromagnetic radiation may be tuned for selective absorption by a particular isotopologue, or it may be from a broadband source.

[0015] In an alternative embodiment the system is coupled to the input of a centrifuge or other mass separation device. The application of electromagnetic radiation and/or magnetic fields to a gas flowing through the reactor provides for a transient enhancement of the mass difference between chemical species bearing two distinct isotopes. The transient enhancement of the mass difference has a lifetime that may be greater than the time required for passage through the mass separation device.

BRIEF DESCRIPTION OF THE DRAWINGS

[0016] FIG. 1A shows a block diagram of an electrolytic cell interphase control system in accordance with an embodiment of the present invention.

[0017] FIG. 1B shows a schematic diagram of a parallel driver module coupled to a parallel power module by a tunable delay module in accordance with an embodiment of the present invention.

[0018] FIG. 2A shows a top perspective view of a switched coaxial transmission line bus in accordance with an embodiment of the present invention.

[0019] FIG. 2B shows a top view of the switched coaxial transmission line of FIG. 5A.

[0020] FIG. 2C shows a cross-section view of a switched coaxial transmission line with an attached anode/cathode assembly in accordance with an embodiment of the present invention.

[0021] FIG. 2D shows a bottom perspective view of the switched coaxial transmission line of FIG. 2C.

[0022] FIG. 3A shows an exploded view a parallel plate anode/cathode assembly in accordance with an embodiment of the present invention.

[0023] FIG. 3B shows an assembled view of the parallel plate anode/cathode assembly of FIG. 3A.

[0024] FIG. 3C shows a cross section view of a parallel plate anode/cathode assembly in accordance with an embodiment of the present invention.

[0025] FIG. 4A shows an exploded view of a solid dielectric coaxial anode/cathode assembly in accordance with an embodiment of the present invention.

[0026] FIG. 4B shows a perspective view of a solid dielectric coaxial anode/cathode assembly attached to a parallel plate transmission line in accordance with an embodiment of the present invention.

[0027] FIG. 5 shows a perspective view of a liquid dielectric coaxial anode/cathode assembly attached to a parallel plate transmission line in accordance with an embodiment of the present invention.

[0028] FIG. 6A shows a perspective view of a transmission line duct with opposing anode and cathode walls in accordance with an embodiment of the present invention.

[0029] FIG. 6B shows a perspective view of a transmission line duct with opposing anode and cathode walls with integrated switches in accordance with an embodiment of the present invention.

[0030] FIG. 6C shows a perspective view of a transmission line duct with opposing anode and cathode walls with a detachable switch module in accordance with an embodiment of the present invention.

[0031] FIG. 6D shows a perspective view of a transmission line duct with opposing anode and cathode walls with a detachable switch module in an attached configuration in accordance with an embodiment of the present invention.

[0032] FIG. 7A shows a perspective view of a dual duct transmission line with an integrated power supply in accordance with an embodiment of the present invention.

[0033] FIG. 7B shows a perspective view of a dual duct transmission line with an integrated power supply and dual switching in accordance with an embodiment of the present invention.

[0034] FIG. 7C shows a perspective view of an illuminated transmission line duct coupled to an electrolyte circulation system in accordance with an embodiment of the present invention.

[0035] FIG. 8A shows a perspective view of a parallel plate transmission line anode/cathode assembly on a dielectric substrate in accordance with an embodiment of the present invention.

[0036] FIG. 8B shows a top view of the parallel plate anode/cathode assembly of FIG. 13A.

[0037] FIG. 9 shows an electrolytic cell power supply output waveform in accordance with an embodiment of the present invention.

[0038] FIG. 10 shows an electrolytic cell power supply circuit schematic diagram in accordance with an embodiment of the present invention.

[0039] FIG. 11 shows a schematic diagram of an output stage of an electrolytic cell power supply in accordance with an embodiment of the present invention.

[0040] FIG. 12A shows a schematic diagram of a complementary output circuit for driving an electrolytic cell with dual voltages in accordance with an embodiment of the present invention.

[0041] FIG. 12B shows a schematic diagram of a complementary output circuit for driving an electrolytic cell with four voltages in accordance with an embodiment of the present invention.

[0042] FIG. 13A shows an electrode assembly with a transparent electrode and magnet in accordance with an embodiment of the present invention.

[0043] FIG. 13B shows an electrode assembly with a transparent window and coplanar electrode assembly in accordance with an embodiment of the present invention.

[0044] FIG. 14 shows a coaxial transmission line with a circulating center conductor in accordance with an embodiment of the present invention.

[0045] FIG. 15A shows an electrolytic cell with a circulating center conductor in accordance with an embodiment of the present invention.

[0046] FIG. 15B shows a coaxial transmission line with multiple electrolytic cells in a series configuration in accordance with an embodiment of the present invention.

[0047] FIG. 16 shows a coaxial transmission line with a liquid metal electrode in accordance with an embodiment of the present invention.

[0048] FIG. 17A shows a stabilized liquid metal electrode in accordance with an embodiment of the present invention.

[0049] FIG. 17B shows a coaxial transmission line with a series configuration of stabilized liquid metal electrodes in accordance with an embodiment of the present invention.

[0050] FIG. 18A shows a perspective view of a coaxial electrolytic cell module in accordance with an embodiment of the present invention.

[0051] FIG. 18B shows a cutaway view of the coaxial electrolytic cell of FIG. 18A.

[0052] FIG. 18C shows a section view of the electrolyte ports axis of the coaxial electrolytic cell of FIG. 18A.

[0053] FIG. 18D shows a section view of the liquid metal ports axis of the coaxial electrolytic cell of FIG. 18A.

[0054] FIG. 19 shows a schematic for an RC time constant measurement circuit in accordance with an embodiment of the present invention.

[0055] FIG. 20 shows a schematic for a redox reaction detection circuit in accordance with an embodiment of the present invention.

[0056] FIG. 21A shows a schematic for an electrolytic redox circuit with a high frequency source in accordance with an embodiment of the present invention.

[0057] FIG. 21B shows a schematic for a transmission line duct with a pulsed excitation source in accordance with an embodiment of the present invention.

[0058] FIG. 22A shows a perspective view of a parallel plate transmission line duct with a shunt switch in accordance with an embodiment of the present invention.

[0059] FIG. 22B shows an exploded view of the parallel plate transmission line duct shown in FIG. 22A.

[0060] FIG. 22C shows a section view of the parallel plate transmission line duct shown in FIG. 22A.

[0061] FIG. 22D shows a section view of a parallel plate transmission line duct with solid electrodes in accordance with an embodiment of the present invention.

[0062] FIG. 23 shows a perspective view of a parallel plate transmission line duct with a coupled single turn solenoid in accordance with an embodiment of the present invention.

[0063] FIG. 24 shows a flow diagram for an isotope separation process in accordance with an embodiment of the present invention.

[0064] FIG. 25A shows a timing diagram for an isotope separation process with non-overlapping pulses and a simple extraction pulse in accordance with an embodiment of the present invention.

[0065] FIG. 25B shows a timing diagram for an isotope separation process with non-overlapping pulses and a complex extraction pulse in accordance with an embodiment of the present invention.

[0066] FIG. 25C shows a timing diagram for an isotope separation process with overlapping magnetic excitation and extraction pulses in accordance with an embodiment of the present invention.

[0067] FIG. 25D shows a timing diagram for an isotope separation process with overlapping photolytic and extraction pulses in accordance with an embodiment of the present invention.

[0068] FIG. 26 shows a schematic for an electrolytic cell with a magnetic excitation source in accordance with an embodiment of the present invention.

[0069] FIG. 27 shows an electrolytic cell with a magnetic excitation transmission line in accordance with an embodiment of the present invention.

[0070] FIG. 28 shows an exploded view of the electrolytic cell of FIG. 27.

[0071] FIG. 29 shows an exploded view of the magnetic excitation transmission line of FIG. 27.

[0072] FIG. 30 shows a photolytic transmission line duct in accordance with an embodiment of the present invention.

[0073] FIG. 31 shows a photolytic transmission line duct coupled to a mass effect isotope separation device in accordance with an embodiment of the present invention.

[0074] FIG. 32 shows a serial transmission line duct reactor in accordance with an embodiment of the present invention.

[0075] FIG. 33 shows a magnetic excitation transmission line in accordance with an embodiment of the present invention.

[0076] FIG. 34 shows a magnetic excitation transmission line coupled to a coaxial transmission line in accordance with an embodiment of the present invention.

[0077] FIG. 35 shows an embedded magnetic excitation transmission line reactor in accordance with an embodiment of the present invention.

DETAILED DESCRIPTION OF THE INVENTION

[0078] FIG. 1A shows a block diagram 100 of an embodiment of an electrolytic cell interphase control system. A bus 111 couples a control module 110 to a waveform generator 115. The control module 110 transmits signals to the waveform generator 115 that set the parameters of an output waveform (e.g., duty cycle, amplitude, and period). The bus 111 may also provide feedback to the control module 110 with respect to the output of the waveform generator 115. The output waveform of the waveform generator 115 may include a unipolar signal that has a positive excursion referenced to ground and/or a bipolar signal with positive and negative excursions.

[0079] The waveform generator 115 is coupled to a driver 120 by a signal bus 112. The bus 112 may couple two nodes

and carry a single waveform as the output of the waveform generator 115, or it may carry a number of distinct signals between more than two nodes. In a preferred embodiment the driver 120 is driven by an input signal in the range of 1-10 volts and has output rise and fall times of less than 50 nanoseconds. The driver 120 is coupled to the control module 110 by a bus 125 that allows the control module 110 to monitor the driver output and/or control the supply voltage for the driver 120.

[0080] The driver 120 is coupled to a redox power module 130 that is essentially a switched current supply that provides current to an electrode assembly 140 via a transmission line 135. The power module may include N-channel and/or P-channel MOSFETs (metal-oxide semiconductor field-effect transistors). In a preferred embodiment the power module includes multiple selectively switched MOSFETs coupled to three or more supply voltages. The power module 130 is coupled to the control module 110 by bus 125, allowing for control of the supply voltages to the MOSFETs. A bus 127 may be used to provide feedback to the control module 110 from the electrode assembly 140.

[0081] In addition to MOSFETs, JFETs (junction field effect transistors), BJTs (bipolar junction transistors), and IGBTs (insulated-gate bipolar transistors) may be used as switches in the power module 130. Generally, the turn-off speed of silicon BJTs and IGBTs is inferior to that of silicon MOSFETs. However, BJTs using materials such as gallium arsenide and indium phosphide and employing heterojunction structures can provide considerable improvements over silicon BJTs. JFETs may be preferred for low voltage applications.

[0082] The transmission line 135 is preferably a coaxial transmission line or a parallel plate transmission line, or may be a combination of the two. In a preferred embodiment, the gap between conductors in the transmission line is substantially filled with a solid dielectric. It is desirable that the two conductors be restrained from moving under the influence of the magnetic fields generated by the current flowing through them. If the two conductors are able to respond to the magnetic fields that are generated, they may act as an electromechanical transducer that presents a variable load to the power module 130, thus altering the waveform at the electrode surface. For coaxial conductors, a displacement of the axis of the center conductor with respect to the axis of the outer conductor does not affect the DC inductance; however, it can affect the inductance at high frequencies.

[0083] For purposes of this disclosure, a statically configured transmission line is defined as a restrained pair of conductors configured as a transmission line with a sufficiently small spacing between them such that if they were not restrained, one or both conductors would experience a displacement as a result of the electromagnetic force generated by an operational current flowing through the pair of conductors. Operational current is defined as a current that would flow through the conductors during normal operation.

[0084] The electrode assembly 140 is preferably a transmission line structure, with the anode and cathode serving as the two conductors in the transmission line in contact with electrolyte 145. In one embodiment, the gap between the anode and cathode is substantially filled with a solid dielectric. In another embodiment, the gap between the anode and cathode is substantially filled with electrolyte 345. Frequent reference will be made in this specification to an "electrode assembly" or an "anode/cathode assembly" with two elec-

trodes. Unless specifically stated otherwise, either of the two electrodes may serve as anode or cathode, with a reference to one designation implying the substitution of the other as an alternative embodiment.

[0085] For purposes of this disclosure, an “electrode” is a conductor that is intended to be used in contact with an electrolyte, and may be either an anode or a cathode. A “bus” is a conductor that may be used to couple an electrode to a power source or signal source, but is itself not intended to be used in contact with an electrolyte. A “transmission line” may refer to either a parallel plate transmission line or a coaxial transmission line.

[0086] For purposes of this disclosure, in reference to a parallel plate transmission line, a preferred but not exclusive embodiment thereof is a pair of substantially flat rectangular conductors that have a spacing s and a width w such that the inductance per unit length L in Henries/meter is approximated by the equation:

$$L = 4\pi \times 10^{-7} \left(\frac{s}{w} \right)$$

[0087] In general, there are a number of spatial arrangements of conductors that can be used for transmission lines, such as parallel wires, parallel plates, and coaxial conductors. For purposes of this disclosure, in reference to a transmission line, a preferred but not exclusive embodiment thereof includes a spatial arrangement of conductors that is mechanically fixed to maintain the spatial arrangement under load.

[0088] Electrolyte **145** may be an aqueous or nonaqueous solvent containing dissolved ions. A nonaqueous solvent may be an aprotic solvent. The electrolyte **145** may include one or more molten salts such as an alkali metal fluoride or chloride. Electrolyte **145** may also include an ionic material that is a liquid at room temperature. In contrast to electrochemical energy storage devices, which may have closely spaced planar electrodes, the volume of electrolyte **145** in contact with the electrode assembly **140** is typically larger than the volume between the electrodes. An electrolytic cell that is used for a manufacturing process requires access to reactant species to replace those converted to product species.

[0089] Electrolyte **145** may contain isotopically enhanced reactant species. For example, an aqueous electrolyte may use heavy water (D_2O) or an organic acid may include ^{13}C at a concentration that is above that found in nature. The selection of electrolyte species enhanced with magnetic or nonmagnetic isotopes provides for additional control of reactions in the interphase through spin locking or spin conversion.

[0090] For purposes of this disclosure, the term “accessible electrolyte volume” refers to the volume of electrolyte in an electrolytic cell that is in electrical contact with the anode and cathode. In a preferred embodiment for parallel plate or coaxial transmission line electrode assemblies, the accessible electrolyte volume is at least ten times greater than the volume swept out by the projection of one electrode onto the other.

[0091] A sensor **150** is in contact with the electrolyte **145** and coupled to the Control Module **110** by bus **126**. Sensor **150** may be a reference electrode, temperature sensor or resistance measurement cell. Sensor **150** provides information feedback for process control by the Control Module **110**. Sensor **150** may provide information concurrent with the

output of power module **130**, or the output of power Module **130** may be suspended while Sensor **150** is operational.

[0092] A high frequency current source **155** is coupled to the transmission line **135** and is also coupled to the control module **110** by a bus **128**. The high frequency current source produces a magnetic field that permeates a portion of the electrolyte **145** and thus may be used to alter the spin evolution of species within the electrolyte **145**. The current from the high frequency current source may flow in one or both electrodes of the electrode assembly **140**.

[0093] In an embodiment the control module **110** is used to control an array of electrode assemblies **140** with each electrode assembly coupled to a dedicated high frequency current source **155**. Each electrode assembly may also be coupled to a dedicated redox power module **130**. The array of electrode assemblies **140** may be integrated on a single substrate (e.g., silicon). For spin locking or spin conversion of reactant species, the high frequency current source may operate at microwave frequencies (e.g., >1 GHz). In order to minimize losses and reduce switching transients, an array of individually driven small area electrode assemblies (e.g., <1 cm²) is preferred.

[0094] FIG. 1B shows a schematic diagram of a parallel driver module **120a** coupled to a parallel power module **130a** by a tunable delay module **122**. Driver1 is coupled to switch1 by a delay element delay1 and driver2 is coupled to switch2 by a delay element delay2. In order to obtain high currents, parallelism among drivers and switches may be required. Multiple individual driver circuits may be combined as driver1 or driver2. For example, both driver circuits on a dual integrated circuit (IC) may be combined to drive a single transistor. Although less common, more than one switch circuit may be combined to be driven by a single driver. When a large number of driver/switch combinations are combined in parallel, variation in switching behavior between driver/switch combinations will tend to degrade the output waveform.

[0095] The delay module **122** provides a tunable delay1 between driver1 and switch1 and a tunable delay2 between driver2 and switch2. For switches with logic level inputs (e.g., logic level input MOSFETs) a monostable multivibrator such as the 74VHC221A device manufactured by the Fairchild Semiconductor Corporation may be used. For switches requiring a high drive voltage, the MM74C221 monostable multivibrator from the Fairchild Semiconductor Corporation may be used. The delay may be tuned once during manufacturing, or it may be tuned periodically during operation. For operational tuning, a digital potentiometer such as the AD5222 manufactured by Analog Devices, Inc. may be used to set the RC time constant for a monostable multivibrator.

[0096] Delay1 and/or delay2 may be adjusted to minimize the distortion in the output waveform. Although only two driver/delay/switch combinations are shown, several may be used in an electrolytic cell interphase control system. In general, the greater the number of switches (e.g., transistors) configured in parallel, the greater the benefit of tunable delays. In a preferred embodiment the output rise and fall times of the power module **130a** are less than 100 nanoseconds.

[0097] FIG. 2A shows a top perspective view **200** of an embodiment of a switched coaxial transmission line bus **205**. An outer conductor **210** encloses a dielectric **215**, which in turn encloses a center conductor **220**. Switches **230** couple center conductor **220** to conductor plate **225**.

[0098] FIG. 2B shows a top view 201 of the switched coaxial transmission line of FIG. 2A. For clarity, dielectric 215 and outer conductor 210 are shown cut back to expose a portion of center conductor 220 and conductor plate 225. The exposed surface of conductor plate 225 may be used to mount circuit elements associated with the switches 230. In practice, dielectric 215 and outer conductor 210 may envelop a greater area of conductor plate 225 and inner conductor 220. FIG. 2C shows a cross-section view 202 of the switched coaxial transmission line bus 205 of FIG. 2A with an integrated anode/cathode assembly including planar electrode 235 and planar electrode 240.

[0099] FIG. 2D shows a bottom perspective view 203 of the switched coaxial transmission line of FIG. 2C. Planar electrode 240 and planar electrode 235 are shown without a dielectric. However, it should be noted that both filled and unfilled coaxial and parallel plate transmission lines may be used as anode/cathode assemblies and attached to coaxial and parallel plate transmission line busses using an orthogonal transition.

[0100] FIG. 3A shows an exploded view 300 of an embodiment of a parallel plate anode/cathode assembly. First and second mounting blocks 305 and 310 are provided for establishing an electrical connection to electrodes 325 and 315, respectively. Mounting blocks 305 and 310 also provide a means for attaching the assembly to a parallel plate or coaxial transmission line bus.

[0101] Electrodes 325 and 315 are separated by a dielectric 320. The dielectric 320. Copper is a preferred material for electrodes 325, which may be coated with other metals (e.g., platinum) to provide a working surface with different properties. If a high permeability material such as nickel is used as a coating, it is desirable that the coating be kept thin to avoid an undue increase in inductance. The dielectric 320 may be a ceramic, a polymer, or a composite material. It may also be a sheet form that is bonded to electrodes 325 and 315. Alternatively, it may be a dielectric adhesive that is applied to electrode 325 and/or electrode 315.

[0102] FIG. 3B shows an assembled view 301 of the parallel plate anode/cathode of FIG. 3A. The hole 306 provides a path for current and mass transport between the two electrode surfaces. More than one hole may be provided, depending upon the desired current distribution and mass transport between the electrode surfaces. The parallel plate transmission line with a solid dielectric and the parallel plate electrode with a gap can be viewed as the opposite ends of a spectrum of parallel plate electrode configurations, with perforated solid dielectric parallel plate electrodes falling in between. In one embodiment, the dielectric is a ceramic substrate and electrode 315 and 320 are deposited using thin film or thick film techniques such as those used for electronic circuits. Hole patterns in the ceramic substrate may be punched in green tape before firing.

[0103] FIG. 3C shows a cross section view 302 of an embodiment of a parallel plate anode/cathode assembly. An anode 330 and a cathode 335 are bonded together by a thermosetting polymer adhesive 340 that contains filler particles 345. The thermosetting polymer adhesive may be an epoxy and the filler particles may be silica, alumina, or other ceramic.

[0104] FIG. 4A shows an exploded view 400 of a solid dielectric coaxial anode/cathode assembly. A coaxial element 490 includes center electrode 405 and an outer electrode 415 that are separated by a dielectric 410. The cutback of the outer

conductor 415 at the upper end provides a more uniform current distribution at the electrode surfaces. A first plate conductor 425, a dielectric 430, and a second plate conductor 435 make up a socket portion 491 of a parallel plate transmission line for connection to the coaxial element 401.

[0105] FIG. 4B shows a perspective view 401 of a solid dielectric coaxial anode/cathode assembly 490 attached to a switched coaxial transmission line 445.

[0106] Switched coaxial transmission line 445 is similar to switched coaxial transmission line 205 shown in FIG. 2A. It should be noted that the orthogonal transition shown in FIGS. 4A and 4B may be used to connect a circular coaxial transmission line bus to a parallel plate transmission line electrode assembly. In practice, it is usually more straightforward to use a parallel plate transmission line or a coaxial transmission line with a rectangular cross-section since the switched current source typically has a planar layout to begin with.

[0107] FIG. 5 shows a perspective view 500 of a coaxial anode/cathode assembly 590 attached to parallel plate transmission line 505. The coaxial anode/cathode assembly 590 has a center electrode 510 separated from an outer electrode 515 by a gap 520.

[0108] FIG. 6A shows a perspective view 600 of a transmission line duct 691 with an anode wall 615 and an opposing cathode wall 620. As with other transmission line conductors discussed herein, the anode wall 615 and cathode wall 620 are preferably fabricated from a material with high electrical conductivity and low magnetic permeability such as copper. For applications where dimensional stability is desired, particularly at high temperatures, tungsten or molybdenum may be used. The base electrode may be coated with another metal to obtain particular surface characteristics. For a particular electrolyte, the surface coating may be chosen to provide a non-polarizable or low polarization electrode. Anode wall and/or cathode wall 620 may be partially masked to provide a desired ratio between the active areas of anode wall 615 and cathode wall 620.

[0109] A first dielectric wall 625 and a second dielectric wall are sandwiched between the anode wall 615 and the cathode wall 630, and their height determine the height of the duct channel 635. Dielectric wall 625 and 630 are preferably fabricated from a dielectric material that is inert with respect to the electrolyte contemplated for use. For very short walls, a stiff, creep resistant material such as silica, alumina, beryllia, or other ceramic is preferred to maintain dimensional stability. Non-oxide ceramics such as silicon nitride, boron nitride, silicon nitride, and aluminum nitride may be used. The dielectric and conductive structures may also be integrated on a semiconductor substrate.

[0110] Top backup plate 605 and bottom backup plate 610 are not required, but are preferred when the anode wall 615 and cathode wall 620 are thin and additional mechanical support is desired. The anode wall 610 and the cathode wall 615 may be fabricated on the top backup plate 605 and the bottom backup plate 610, respectively, using thin-film or thick film techniques such as those used for fabricating electronic circuits on ceramic or semiconductor substrates. Patterning may be done using photolithographic techniques. Single crystal and polycrystalline ceramic materials may be lapped and polished to provide backup plates with high dimensional accuracy. Thin gold metallization may be applied along with appropriate adhesion layers to provide

diffusion bondable surfaces. Opaque and/or transparent ceramic materials may be used for backup plate **605** and/or backup plate **610**.

[0111] The anode wall **610** and/or the cathode wall **615** may be fabricated by depositing transparent conductive materials on the top backup plate **605** and the bottom backup plate **610**, respectively. Examples of suitable transparent conductive materials are antimony doped tin oxide and tin doped indium oxide. Transparent conductive materials may be deposited alone or in combination with a fine-line metal pattern for enhanced conductivity. Examples of materials that are suitable for use as top backup plate **605** and bottom backup plate **610** are sapphire and fused silica. For greater transmission in the IR region, sulfides, selenides and halides may be used. The use of transparent materials for the backup wall and anode/cathode walls enables the illumination of the electrode surfaces.

[0112] The flat surface surrounding the duct channel **635** provides an area against which a seal may be made to enable a forced fluid flow through the channel duct **935**.

[0113] Additional backup plates may be added to increase the seal surface area around the channel duct **635**. A temporary seal may be made using gaskets or o-rings, and a more permanent seal may be made using adhesives. The use of ceramic materials and thin film techniques enables the construction of ducts with a height on the order of 0.001 inches or smaller and a width on the order of an inch or larger. For low profile transmission line ducts, adapters may be attached to facilitate plumbing connections. The transmission line duct **691** is an embodiment of a fundamental element of the present invention: an electrolytic cell with inherently low inductance that is achieved through closely spaced and substantially parallel electrodes with a separation that is small compared to the width of parallel plate electrodes. A transmission line duct with coaxial electrodes will have an electrode separation that is small in comparison to the cross-section perimeter of the center conductor. In a preferred embodiment of transmission line duct **691**, the width to separation ratio of the anode wall **610** and the cathode wall **615** is at least 100.

[0114] FIG. 6B shows a perspective view **601** of an embodiment of a switched transmission line duct **692**. An anode wall **616** and a cathode wall **621** are coupled to and separated by a dielectric wall **631** and a transmission line dielectric **626**. The transmission line dielectric **626** is also coupled to a switch plate **617** and separates switch plate **617** from the cathode wall **621**. The switch plate **617** is coupled to anode wall **616** by switches **640** (e.g., transistors). The switched transmission line duct **692** is essentially a union of two parallel plate transmission lines, with two of the conductors being directly coupled and the other two conductors being coupled by switches.

[0115] FIG. 6C shows a perspective view **602** of an embodiment of a transmission line duct **693** with a detachable switch module **694**. The transmission line duct **693** is similar to the transmission line duct **691** of FIG. 6A, but has been adapted for detachable coupling to the power module **694**. Dielectric walls **631a** and **631b** are disposed between anode wall **618a** and cathode wall **613a**, which are in turn sandwiched between backup plate **607a** and backup plate **612a**.

[0116] The detachable switch module **694** has a lower conductor plate **613b** and an upper conductor plate **618b** that are separated by and coupled to a transmission line dielectric **631c**. The transmission line dielectric **631c** is also coupled to a switch plate **619** and separates switch plate **619** from the

lower conductor plate **613b**. The switch plate **619** is coupled to upper conductor plate **618b** by switches **640** (e.g., transistors).

[0117] FIG. 6D shows a perspective view **603** of the transmission line duct **693** and detachable power module **694** of FIG. 6C in an attached configuration. Dielectric wall **631b** and transmission line dielectric **631c** interlock as a tongue-in-groove. Anode wall **618** overlaps upper conductor plate **618b**, and cathode wall **613a** overlaps bottom conductor plate **613b**. The detachable power module is desirable when an array of transmission line ducts **693** are arranged in the same fluid electrolyte circuit. If a power module **694** fails, it can be replaced without disturbing the fluid electrolyte circuit.

[0118] In an electrolytic cell with an aqueous electrolyte, a nominal double-layer capacitance of 20 microfarads per square centimeter and an electrode area of 25 square centimeters, the average current required to charge the capacitance to one volt in one microsecond is on the order of 500 amperes. Faster charging times will require proportionally larger currents, with peak currents on the order of thousands of amperes.

[0119] For an electrolytic manufacturing process that requires large total electrode areas in order to obtain a reasonable throughput, driving a single large electrolytic cell (e.g., plating bath) will be very difficult. Thus, it is an aspect of the present invention to provide a compact module that combines an electrolytic cell with a local power supply.

[0120] Another aspect of the invention is the combination of an array of compact modules to provide a large total electrode area.

[0121] The inductance of a circuit element increases with length. It is thus desirable to minimize the circuit path between the switch and the anode/cathode of a high-speed electrolytic cell. Instead of increasing the size of a power supply and the electrolytic cell it serves, the electrolytic cell can be divided into a plurality of smaller cells, each with a dedicated power supply. To reduce the overall load capacitance and thus reduce the peak current, an array of electrolytic cells may be configured in series. The smaller capacitance will reduce the charging current that is required; however, the overall applied voltage will be increased.

[0122] FIG. 7A shows a perspective view **700** of an embodiment of an electrolytic module **790** that is derived from the transmission line duct **692** shown in FIG. 6B. The duct channel **636** of transmission line duct **692** has been subdivided by a septum **710** to produce two adjacent duct channels **705a** and **705b**. Septum **710** may be used to provide additional dimensional stability and accuracy for closely spaced anodes and cathodes. A control circuit board **715** has also been added. Vias may be used to connect circuit elements on the circuit control board **715** to the transmission line conductors.

[0123] Control circuit board **715** provides a number of control functions for the switch transistors **720a**, **720b**, and **720c**. Bypass capacitors **725a**, **725b**, and **725c** are in close proximity to switch transistors **725a**, **725b**, and **725c**, and serve to minimize voltage drops at turn-on. Bypass capacitors **725a**, **725b**, and **720c** preferably have a low equivalent series resistance. Multiple capacitors may be used in parallel for each transistor. Transistor driver **735** provides the drive signal to switch transistors **720a**, **720b**, and **720c**. Transistor driver **735** may be a MOSFET driver, and more than one may be used to drive the switch transistors **720a**, **720b**, and **720c**. Waveform generator **740** provides the waveform that is amplified by

transistor driver(s) **735**. Voltage regulators **730a**, **730b**, and **730c** provide the supply voltages to switch transistors **720a**, **720b**, and **720c**.

[0124] Microcontroller **745** controls the output voltages of voltage regulators **730a**, **730b**, and **730c**. Microcontroller **745** may have a built-in Analog-to-digital conversion capability that provides for adjustment of the voltage regulators in response to measured I-V characteristics of the anode and cathode. Microcontroller **745** may also have a communications capability that allows it to be networked with a master controller, thus allowing a central master controller to control an array of electrolytic modules **790**. Examples of devices suitable for use as microcontroller **745** are the Z8 Encore!®8K Series of 8-bit microcontrollers manufactured by Zilog, Inc.

[0125] The functions described in relation to circuit board **715** may be provided by different configurations of integrated circuits and discrete devices. Field programmable gate arrays (FPGAs) or application specific integrated circuits (ASICs) may also be used. Additional switch transistors, bypass capacitors, and voltage regulators may be added to provide more complex output waveforms and high frequency excitation.

[0126] FIG. 7B shows a bottom perspective view **701** of an embodiment of a double-switched electrolytic module **791** that is derived from the electrolytic module **790** shown in FIG. 7A. The cathode wall **921** of electrolytic module **790** has been divided into a cathode wall **721a** and a switch plate **721b**, which are coupled by switching transistors **725d**, **725e**, and **725f**. A circuit board **755** is coupled to switch plate **721b**, and serves to drive switching transistors **725d**, **725e**, and **725f**. The use of two sets of switches allows both terminals of the electrolytic cell to be driven, thus enabling the use of a bridge configuration. Circuit board **755** may be configured as a slave circuit, with circuit board **715** serving as a master. In slave mode, circuit board **755** may or may not include a microcontroller and/or waveform generator. Stiffener **760** has been added to prevent the loss mechanical integrity that may occur from the division of the cathode wall. Modifications to the location of the switched gaps and to the backup plates may also be made to improve mechanical integrity.

[0127] FIG. 7C shows a perspective view **702** of an illuminated electrolytic module **792** that is derived from the electrolytic module **790** of FIG. 7A. An input adapter **760** and an output adapter **762** are coupled to a circulation pump **770** by conduits **765** and **767**, respectively. Input adapter **760** and output adapter **762** are coupled to opposite ends of the channel duct of electrolytic module **792** to provide forced flow of electrolyte through the channel duct.

[0128] Illumination module **780** may be provided as a photon source for use with transparent backup plate/electrode assemblies to provide radiation at an electrode surface to assist redox reactions. The illumination module may be a continuous source or it may be a pulsed source. The illumination module may be controlled by the circuit board **715**. As a pulsed source, the illumination module may be synchronized with a switch driver waveform output by the circuit board **715**.

[0129] The illumination module **780** may be a monochromatic light source or a filtered light source for providing a limited spectrum. Light emitting diodes (LEDs) and/or laser diodes may be used as elements in the illumination module **780**. The illumination module **780** may include fiber optics or

other transmission means to couple the electrolytic module **792** to a remote photon source (e.g., a tunable dye laser).

[0130] FIG. 8A shows a perspective view **800** of an embodiment of a parallel plate (microstrip) transmission line **890** including an anode **810**, a dielectric **815** and a cathode **820** on a dielectric substrate **805**. The dielectric substrate **805** may be a ceramic material, or it may be a semiconductor substrate with a dielectric coating such as silicon dioxide or silicon nitride.

[0131] FIG. 8B shows a top view **801** of the parallel plate transmission line **890** of FIG. 12A. The anode **810** is essentially a continuous sheet of conductive material deposited on the surface of dielectric substrate **805**. A deposited dielectric film **815** separates the anode **810** from the cathode **820**. The dielectric **816** has an apron region **816** extending out from the edge of the cathode **820**. The cathode **820** includes an array of fingers **820a** having a width W_2 , adjacent to anode stripes **810a** having a width W_1 . The ratio W_1/W_2 may be varied to adjust the electrode area ratio. For a given substrate area, decreasing W_1 and W_2 decreases the total resistance between the anode **810** and the cathode **820**. The pattern **890** may stepped and repeated over a large area with bus connections to each pattern. W_1 and W_2 may be on the order of a micron.

[0132] Due to a large resistance or a large capacitance, or both, the RC time constant of an electrolytic cell may prevent the voltage across the double-layer capacitance in the cell from rising quickly enough to suit a particular process. In this instance, a voltage greater than the desired working cell voltage may be applied for a short duration to accelerate charging or discharging of the double-layer capacitance.

[0133] FIG. 9 shows a diagram **900** for an embodiment of a waveform applied to an electrolytic cell for control of an interphase in an electrolytic cell. It is important to note that V_{cell} is the voltage applied to the electrolytic cell as a whole. At the beginning of an electrolytic process a voltage V_0 is applied for a period t_0 . The application of V_0 establishes a concentration profile for each of the charged species within the interphase at the electrodes of the electrolytic cell. The length of period t_0 is preferably sufficient for the concentration profiles of the species of interest to equilibrate. V_0 is generally a voltage at which no intended redox reactions occur, although a small net current may be observed due to redox reactions involving impurities. Although V_0 is shown to be opposite in polarity to V_1 and V_2 , it may be of the same polarity. The waveform of FIG. 9 may be produced by the system shown in FIG. 1A.

[0134] For example, if the intended electrolytic process is a reduction reaction at the cathode, the application of V_0 to the electrode serving as the anode will produce a positive charge at the cathode. This positive charge will lower the cation concentration within the interphase at the cathode surface and increase the anion concentration in the interphase at the cathode surface. The mean distance between the cathode surface and the cations within the interphase will be increased.

[0135] Subsequent to period t_0 , a voltage V_1 is applied for a period t_1 . V_1 is a voltage that is greater in magnitude than the voltage V_2 at which the intended reaction will occur. For systems including a solvent and a dissolved electrolyte, V_1 may be equal to or greater than the cell potential at which the solvent is oxidized and/or reduced. For embodiments in which the electrolyte has a low conductivity, it is preferred that V_1 be greater than the voltage at which solvent electrolysis occurs.

[0136] It is important that V_1 and t_1 are closely controlled, since overcharging of the double-layer capacitance may occur. In processes where V_1 is greater than the voltage at which solvent electrolysis occurs, electrolysis is inevitable if t_1 is not sufficiently limited. The purpose of the (V_1, t_1) pulse is to overcome the RC time constant of the electrolytic cell. Ideally, at the end of t_1 , the potential across the double-layer capacitance is equal to the desired process potential associated with the cell voltage V_2 , and has been reached in a time t_1 that is less than the time it would have taken if V_2 were applied directly.

[0137] The change in polarity from V_0 to V_1 and the magnitude of V_1 may result in large currents during the initial charging of the double-layer capacitance. It is important that the power supply providing V_1 have a low inductance and a low internal resistance so that current lag and limiting are minimized.

[0138] V_2 is the cell voltage at which the desired reaction (e.g., reduction at the cathode) occurs. V_2 may be the voltage associated with the onset of the reaction, but is preferably one hundred millivolts or more higher. Due to the small distances and short timescales involved with the interphase, it is desirable to carry out redox reactions with large overpotentials so that charge transfer kinetics are not a limiting factor. It is preferable that V_2 provide a sufficiently large reaction overpotential so that the time required for migration of a cation to the electrode is large compared to the time required for its reduction.

[0139] During the application of (V_1, t_1) and (V_2, t_2) , cations will migrate toward the cathode, and their velocity will be influenced by charge, mass, and solvation. Not all cations will have the same velocity under the influence of the applied voltage, thus there will be a degree of segregation between the cations. Segregation may occur between cations with the same mass and different charge, or between cations with the same charge and different mass. The first species to arrive at the cathode will tend to be those with the greatest mobility. The period t_2 may be ended shortly after the first reduction reactions occur, thus limiting reaction participation to the initially closer and faster cations.

[0140] At the end of period t_2 a voltage V_3 is applied for a period t_3 . The purpose of V_3 is to quickly remove the charge acquired by the double-layer capacitance during the application of V_1 and V_2 . This charge removal helps to reset the electrolytic cell so that another pulse cycle can be applied. The application of V_3 for the period t_3 may be omitted from the waveform; however, the discharge of the double-layer capacitance may require a longer time. For processes involving the application of a series of pulses, the (V_3, t_3) segment may be used to increase the pulse rate, and thus the throughput of the process.

[0141] At the end of period t_3 voltage V_4 is applied for a period t_4 . In this instance, V_4 is shown as being different from V_0 ; however, V_4 may be equal to V_0 . In the application of a series of pulses, the (V_0, t_0) segment may be absent altogether (e.g., $V_0=0$). In addition, V_4 is shown as being of opposite polarity from V_1 and V_2 ; however, V_4 may be of the same polarity as V_1 and V_2 . V_4 serves as a reference voltage at which the electrolytic cell is allowed to equilibrate before the next application of V_1 . In one embodiment, the period t_4 is at least ten times greater than the sum of t_1 and t_2 . In another embodiment, the period t_4 is at least 100 times greater than the sum of t_1 and t_2 . Since cation diffusion can be significantly slower than cation migration in a large electric field, a rela-

tively long period may be required for the equilibrium concentration of the cationic species being reduced to be restored in the interphase and the adjacent region in the bulk electrolyte.

[0142] FIG. 10 shows a block schematic view 1000 of an embodiment of an electrolytic cell power supply. A cascade of monostable multivibrators MMV1, MMV2, MMV3, MMV4, MMV5, MMV6, MMV7, and MMV8 form a tapped ring oscillator in which each of the multivibrators MMV1, MMV2, MMV3, MMV4, MMV5, MMV6, MMV7, and MMV8 produces an output pulse with a length that is determined by the time constants R_1C_1 , R_2C_2 , R_3C_3 , R_4C_4 , R_5C_5 , R_6C_6 , R_7C_7 , and R_8C_8 , respectively. The output pulse of MMV1 provides a delay between the output pulses from MMV8 and MMV2 to avoid shootthrough in the NFETs. The output pulse of MMV2 drives a first high input and a first low input of H-bridge driver 1. The output pulse of MMV3 provides a delay between output pulse from MMV2 and MMV4 to avoid shootthrough in the NFETs. The output pulse of MMV4 drives a second high input and a second low input of H-bridge driver 1. An example of a monostable multivibrator suitable for use in the ring oscillator is the TC7WH7123FU from the Toshiba Corporation.

[0143] The output pulse of MMV5 provides a delay between the output pulses from MMV4 and MMV6 to avoid shootthrough in the NFETs. The output pulse of MMV6 drives a first high input and a first low input of H-bridge driver 2. The output pulse of MMV7 provides a delay between output pulse from MMV6 and MMV8 to avoid shootthrough in the NFETs. The output pulse of MMV8 drives a second high input and a second low input of H-bridge driver 1.

[0144] A first pair of outputs of H-bridge driver 1 drives high side NFET5 and low side NFET4. A second pair of outputs of H-bridge driver 1 drives high side NFET3, high side NFET7, and low side NFET4. A first pair of outputs of H-bridge driver 2 drives high side NFET8 and low side NFET1. A second pair of outputs of H-bridge driver 2 drives high side NFET6, high side NFET8, and low side NFET1.

[0145] The circuit of FIG. 10 can be turned on and off by TTL level signals at the enable and trigger input. One or more of resistors R_1 , R_2 , R_3 , R_4 , R_5 , R_6 , R_7 , and R_8 may be digitally controlled potentiometers to allow for altering the pulse width of the monostable multivibrators by a digital signal (e.g., from a microcontroller). Digital potentiometers frequently have a parasitic capacitance, and it must be taken into account when selecting the value for the timing capacitors C_1 , C_2 , C_3 , C_4 , C_5 , C_6 , C_7 , and C_8 . An example of a digital potentiometer suitable for use is the AD5222 Dual Digital Potentiometer manufactured by Analog Devices, Inc.

[0146] H-bridge driver 1 and H-bridge driver 2 in FIG. 10 may be configured from combinations of high side and low side MOSFET drivers. A bridge driver circuit is preferred as a driver for MOSFETs since the use of PFETs can be avoided, allowing all of the output MOSFETs to be NFETs. A Floating gate drive for the high side NFETs allows the supply voltages V_{cc1} , V_{cc2} , V_{cc3} , and V_{cc4} to be significantly larger than the gate-to-source voltage (V_{gs}) on the high side NFETs. V_{gs} is typically less than or equal to 15 volts.

[0147] FIG. 11 shows an electrical schematic diagram 1600 of an embodiment of an NFET output stage of an electrolytic cell power supply. Transistors M1, M2, M3, M4, M5, M6, M7, and M8 correspond to NFET1, NFET2, NFET3, NFET4, NFET5, NFET6, NFET7, and NFET8, respectively of schematic diagram 1400. A load circuit consisting of C1=2 micro-

farads, R1=one megohm, and R3=5 ohms represents an arbitrary load model of an electrolytic cell. An inductance of L1=20 nanohenries is in series with the electrolytic cell. The value for R1=one megohm represents a leakage current. The model is intended to illustrate the charging and discharging behavior of C1, and no appreciable redox reactions are involved.

[0148] Low side NFETs M1 and M4 are driven by sources V3 and V5 respectively. High voltage NFETs M5 and M2 are driven by sources V7 and V9, respectively. Low voltage NFETs M7 and M3 are driven by source V10. NFETs M7 and M3 are configured back-to-back to prevent diode conduction when M5 is on. Similarly, Low voltage NFETs M6 and M8 are driven by source V10 and are configured back-to-back to prevent diode conduction when M2 is on. As an alternative, the back-to-back NFET combination could be replaced by a NFET in series with an external diode at the expense of the diode forward voltage drop.

[0149] FIG. 12A shows an electrical schematic diagram 1200 of an embodiment of a dual voltage complementary MOSFET circuit for driving an electrolytic cell 1203. A positive supply voltage +Vcc is provided to a NFET M1 and a negative supply voltage -Vcc is provided to a PFET M2. The source of M1 and M2 are coupled to one terminal of the electrolytic cell 1203 and the other terminal of the electrolytic cell 1203 is connected to ground. A drive waveform is provided to the gates of M1 and M2 by sources V1 and V2, respectively. M1 and M2 may be augmented by additional transistors M1 and M2 in parallel.

[0150] FIG. 12B shows an electrical schematic diagram 1201 of an embodiment of a quadruple voltage complementary MOSFET circuit for driving an electrolytic cell 1225. Gain block 1205 includes a complementary bipolar transistor pair C1 as a driver stage for NFET M1, which switches high supply +V_{cc2} to the load 1225. Gain block 1210 includes a complementary bipolar transistor pair C2 as a driver stage for back-to-back NFET M2 and NFET M3, which switch low supply +V_{cc1} to the load 1225. Gain block 1215 includes a complementary bipolar transistor pair C3 as a driver stage for PFET M4, which switches high supply -V_{cc4} to the load 1225. Gain block 1220 includes a complementary bipolar transistor pair C4 as a driver stage for back-to-back PFET M5 and PFET M6, which switch low supply +V_{cc3} to the load 1225. The circuit shown in FIG. 15B may be used to provide the waveform shown in FIG. 9.

[0151] FIG. 13A shows an embodiment of an electrode assembly 1300 with a transparent electrode 1310 coupled to an opposing electrode 1305. A photon source 1320 is also coupled to the pulse power supply 1315 and provides emitted electromagnetic radiation 1325 that is transmitted by the transparent electrode 1310 producing transmitted electromagnetic radiation 1330. The transparent electrode 1310 may be fabricated from a transparent substrate such as sapphire or silica coated with a transparent conductive oxide such as indium tin oxide, and/or a patterned metal film. A transparent electrode will generally have greater attenuation than a transparent window that is nonconductive.

[0152] A magnet 1322 may be used to provide a magnetic field in the electrolyte gap between the electrodes. The magnet 1322 may be a permanent magnet or it may be an electromagnet. For high frequency fields an electromagnet with a ferrite core is preferred. The application of a magnetic waveform may be synchronized with the application of the electrical waveform applied to the electrodes (1305, 1310) and

may also be synchronized with the output of the photon source 1320. A static magnetic field may be combined with an alternating magnetic field, and more than one magnet 1322 may be used.

[0153] The magnetic field produced by magnet 1322 is essentially perpendicular to the electrode surface; however, other magnetic field orientations may be employed. For example, a magnet may be oriented so that the magnetic field it produces is parallel to the electrode surface. A static magnetic field may also be oriented from 0 to 90 degrees with respect to an alternating magnetic field produced by currents flowing in the electrodes, or applied independently.

[0154] FIG. 13B shows an embodiment of an electrode assembly 1301 with a transparent window 1311 and an integrated electrode 1306 coupled to a pulse power supply 1315. The integrated electrode 1306 may also be coupled to a RF/microwave current source 1316. Integrated electrode 1306 may be a coplanar interdigitated anode and cathode pattern, or it may be a structure such as that shown in FIG. 8A. A photon source 1320 is also coupled to the pulse power supply 1315 and provides emitted electromagnetic radiation 1325 that is transmitted by the transparent window 1310, producing transmitted electromagnetic radiation 1330. Although not shown, a magnet similar to that shown in FIG. 13A may be used with electrode assembly 1301.

[0155] The integrated electrode assembly 1306 may also serve as a coplanar waveguide or microstrip circuit. The integrated electrode assembly may be fabricated on a dielectric substrate or a semiconductor substrate. When fabricated on a semiconductor substrate, active components such as switches (e.g., a shunt switch) may also be incorporated. For low power systems, the pulse power supply 1315 may also be incorporated on the substrate.

[0156] FIG. 14 shows an embodiment of a circulating coaxial transmission line assembly 1400 having a circulating center conductor 1410. The outer conductor 1405 is separated from the circulating center conductor 1410 by a dielectric 1415. A central duct 1420 and peripheral ducts 1425 provide paths for electrolyte flow to the face of the circulating center conductor 1410. The ducts 1420 and 1425 allow all or a portion of the face of the center conductor to serve as an electrode surface that may be placed in close proximity to an opposing electrode coupled to the outer conductor 1405, while maintaining electrolyte turnover within the gap. The ducts 1420 and 1425 and/or a portion of the center conductor 1410 may include a dielectric material, thereby limiting the active electrode area of the circulating center conductor, or modifying the current path within the circulating center conductor.

[0157] The circulating coaxial transmission line assembly 1400 has a circular cross-section for which the inductance may be approximated by the equation:

$$L = 2 \times 10^{-7} \times \ln\left(\frac{r_o}{r_i}\right)$$

[0158] With respect to the above equation, r_o=inner radius of outer conductor 1405, r_i=outer radius of inner conductor 1410, and L=inductance in henries/meter. Although the circulating coaxial transmission line assembly 1400 is shown with a circular cross-section, other geometries (e.g., rectangular) may also be used.

[0159] FIG. 15A shows an embodiment of an electrolytic cell 1500 that may be used to terminate the circulating coaxial transmission line of FIG. 14. A circulating center conductor 1505 is separated from an outer conductor 1530 by a dielectric 1525. A terminal electrode 1530a is coupled to the outer conductor 1530 and forms an electrolyte gap 1510 in conjunction with the circulating center conductor 1505.

[0160] FIG. 15B shows an embodiment of a coaxial transmission line assembly 1501 having multiple electrolytic cells in a series configuration that may be used in conjunction with the coaxial transmission lines shown in FIGS. 4A and 4B. A center contact electrode 1506 serves as a connection point and the first electrode in the series. Bipolar electrodes 1508 are disposed between the center contact electrode 1506 and a terminal electrode 1531a that is coupled to an outer conductor 1531. Electrodes 1506, 1508, and 1531a are separated by electrolyte gaps 1510, each of which has an intake port 1515 and an exhaust port 1520.

[0161] It is preferable that each electrolyte gap 1510 be served by an independent electrolyte fluid circuit; however, for high resistivity electrolytes a common circuit with a remote common connection may be used. The leakage current due to a common electrolyte connection may be reduced to an acceptable level by maintaining a large resistance between the electrolyte cells 1510 and their common electrolyte connection.

[0162] FIG. 16 shows an embodiment of a circulating electrolytic coaxial transmission line assembly 1600 with a liquid metal electrode 1625. An electrolyte gap 1620 separates the liquid metal electrode 1625 from a circulating center conductor 1605 that is separated from an outer conductor 1615 by a dielectric 1610. The liquid metal electrode 1625 is coupled to the outer conductor 1615. The liquid metal electrode 1625 may be circulated through a circulation reservoir 1630 from which electrolytic reaction products may be extracted by methods such as filtering, centrifuging, cooling, or distillation.

[0163] The liquid metal electrode 1625 may be a metal that is liquid at or near room temperature (e.g., mercury or gallium) and can be used with low melting point electrolytes. For low melting point electrolytes such as room temperature ionic liquids, aqueous electrolytes and organic solvents, polymer materials such as epoxy resins and fluorocarbons may be used in the fabrication of the circulating electrolytic coaxial transmission line assembly 1600. The preferred metals for use in the transmission line assembly are metals that are insoluble in the liquid metal 1625, or metals that form intermetallic compounds with a melting point that is higher than the operating temperature of the circulating electrolytic coaxial transmission line assembly 1600.

[0164] Alternatively, the liquid metal electrode 1625 may be a metal with a higher melting point, thus making it suitable for use with molten halides and other molten salts. Examples of higher melting point metals are: Zn, In, Sn, Sb, Te, Pb, and Bi. The preferred materials for construction of the circulating electrolytic coaxial transmission line assembly 1600 are ceramics such as oxides and nitrides that may be metallized for bonding and providing conductive surfaces. Materials and techniques (e.g., moly-manganese metallized alumina) for metallizing and bonding ceramics that are used in the high power vacuum tube industry are well suited to fabrication of high temperature embodiments of the circulating electrolytic coaxial transmission line assembly 1600.

[0165] A high-melting point liquid metal 1625 may be chosen based on compatibility with a metal that is being reduced. For example, uranium may be reduced from a molten salt electrolyte into a liquid zinc electrode. Metals used in contact with liquid zinc or other liquid metals would preferably be insoluble in liquid metal 1625 or form an intermetallic compound with a melting point that is higher than the operating temperature of the liquid metal 1625.

[0166] FIG. 17A shows an embodiment of a surface-stabilized liquid metal electrode 1700. A container 1705 holds a volume of liquid metal 1710 that may be circulated through an intake port 1715 and an exhaust port 1720. Since many of the electrode configurations associated with embodiments of the present invention may have a small spacing between electrodes, it is desirable that a liquid metal electrode be prevented from developing a short circuit across a small gap. A perforated cover 1725 divides the surface of the liquid metal into a plurality of smaller discrete surfaces 1735. In this embodiment, the cover 1725 includes a material that is not wet by the liquid metal 1710. Although oxides are generally not wet by metals at low temperature, some metals (e.g., gallium on glass) may be able to wet ceramic materials. Nitrides are an alternative to oxides.

[0167] Either the perforated cover 1725 or the container 1705 may be wholly or partly conductive to provide electrical contact to the liquid metal 1710. The cover 1725 may be a flat structure, or may have optional reinforcing features 1730 to provide rigidity. The cover 1725 may be a composite structure that is composed of both dielectric and electrically conductive materials. For example, a metallic base may be coated with a dielectric in those areas that are in contact with an electrolyte. Alternatively, a metallic honeycomb structure may be used to support a thin ceramic plate.

[0168] Forces that may act to destabilize the liquid metal surface include circulation currents in the liquid metal 1710, circulation currents in an electrolyte, and electromagnetic forces due to currents flowing through the electrolytic cell. The division of the metal electrode surface into a plurality of smaller surfaces 1735 increases the force that is necessary to achieve a given displacement of the surface 1735, thus allowing smaller electrolyte gaps to be used in the cell. The smaller electrolyte gaps contribute to lower cell resistance and faster charging of the double-layer capacitance. The viscosity of the electrolyte in contact with the liquid metal 1710 may also be adjusted to dampen oscillations that may arise due to electromagnetic effects.

[0169] FIG. 17B shows an embodiment of an electrolytic coaxial transmission line 1701 with a series configuration of stabilized liquid metal electrodes. The basic unit of the electrolytic coaxial transmission line 1701 is made up of a solid electronic conductor (1745a, 1745, 1740a), an electrolyte-filled gap 1750, and a liquid metal conductor 1760 that are connected in series. The solid electronic conductor 1745a that serves as the first electrode in the first cell is adapted for electrical connection to a power bus. Solid electronic conductors 1745 are adapted to contain and establish electrical contact with the liquid metal 1760. The solid electronic conductor 1740 is adapted to contain the liquid metal 1760 and establish electrical contact to the outer conductor 1740.

[0170] Examples of materials that are preferred for the construction of high-temperature electrodes (1745a, 1745, 1740a) are tungsten/copper and silver/molybdenum composites. These materials have a low magnetic permeability, good electrical conductivity, and their composition can be adjusted

to achieve a good thermal expansion match to a variety of ceramic materials. They can also be coated by a wide variety of other materials to optimize their performance as electrodes and liquid metal containers.

[0171] The outer conductor **1740** is separated from the center conductor elements by dielectric **1775**. Each cell in the coaxial transmission line **1701** has an electrolyte intake port **1765a** and an electrolyte exhaust port **1765b**. Each cell in the coaxial transmission line **1701** also has a liquid metal intake port **1770a** and a liquid metal exhaust port **1770b**. Two different types of liquid metal electrode stabilizing covers are shown. Stabilizing cover **1755** has apertures whose sides are non-wetting with respect to the liquid metal **1760**. Stabilizing cover **1755a** has apertures that are wet by the liquid metal **1760**.

[0172] Stabilizing cover **1755a** is given mechanical support by electrolyte standoff **1780a** and electrode standoff **1780b**. For large area electrodes, standoffs **1780a** and **1780b** stiffen the stabilizing cover and enable the use of smaller electrolyte gaps. Stabilizing cover **1755a** has aperture surfaces that are wet by the liquid metal **1760**, thus providing a liquid metal surface **1756** that is closer to the opposing electrode **1745**.

[0173] FIG. **18A** shows a perspective view of an embodiment of a coaxial electrolytic cell module **1800** that may be used in the construction of a circulating electrolytic coaxial transmission line similar to that shown in FIG. **17B**. A solid electrode **1805a** is separated from an outer electrode **1810** by a dielectric **1815**. Electrolyte ports **1825** and liquid metal ports **1820** are offset **900** from each other. FIG. **18B** shows a cutaway view **1801** of the coaxial electrolytic cell module **1800** of FIG. **18A** and shows a stabilizing liquid metal cover **1830**. FIG. **18C** shows a section view **1802** through the axis of the electrolyte ports **1825**. FIG. **18D** shows a section view **1803** through the axis of the liquid metal ports **1835**. Electrode **1805b** is the counterelectrode to electrode **1805a**.

[0174] FIG. **19** shows a schematic diagram **1900** for an RC time constant measurement circuit that may be used in conjunction with the electrolytic cell interphase control system shown in FIG. **1A**. In order to optimize the waveform provided to an electrolytic cell, it is desirable to know the charging characteristics of the double-layer capacitance associated with the cell. Load **1925** represents an electrolytic cell, or a series of electrolytic cells. Power supply **1930** supplies current to the load **1925** and is controlled by the test controller **1950**. The RC time constant of the load **1925** is measured by determining the time required for discharge of the intrinsic load capacitance from voltage reference V_H to a lower voltage reference V_L .

[0175] In one embodiment, the test controller causes the power supply to charge the load **1925** to a voltage value that is slightly greater than V_H . When charged to a voltage greater than V_H the outputs of comparators **1910** and **1920** are in the same state (e.g., high) since the voltage across the load **1925** is greater than both V_H and V_L . The outputs of comparators **1910** and **1920** are coupled to logic **1935** (e.g., an XOR gate). Clock **1940** is coupled to logic **1935** and a counter **1945** that is enabled to count pulses from clock **1940** when the logic **1935** is in the appropriate state (e.g., XOR high).

[0176] When the test controller **1950** causes the load **1925** to be shunted to ground or other potential that is less than or equal to V_L , the discharge is initiated and the voltage across the load **1925** falls. When the voltage falls below V_H the logic **1935** enables the counting of clock pulses by the counter **1945** until the voltage across the load falls below V_L , at which time

the logic disables the counting of pulses by the counter **1945**. The test controller **1950** may be used to set V_L and V_H so that the RC time constant over a particular voltage range may be determined. The RC time constant thus determined might be used to establish the required pulse width of a fixed voltage pulse that is applied to charge the double-layer capacitance to a desired voltage.

[0177] For example, V_H may be established as the voltage for which the onset of a desired redox reaction occurs in the load **1925**. With respect to FIG. **9** in this instance, V_2 may be a value somewhat larger than V_H . V_L would be set to V_0 or V_4 . The RC time constant for the cell that is determined by the discharge from V_H to V_L may then be used to determine the values for V_1 and t_1 that may quickly charge the double-layer capacitance to V_H ; and also to determine the values for V_3 and t_3 that may quickly discharge the double-layer capacitance.

[0178] FIG. **20** shows a schematic diagram **2000** of an embodiment of a redox reaction detection circuit for detecting the onset of a redox reaction in an electrolytic cell represented by a load **2010**. The circuit may be implemented for real-time control of the potential applied to an electrolytic cell. A power switch **2005** is driven by a driver **2045** that is in turn controlled by output from a logic circuit **2035**. The logic circuit **2035** is responsive to a primary signal **2040** and a comparator **2030**. The comparator **2030** has one output state associated with the condition V_1 greater than or equal to V_2 , and another state associated with V_1 less than V_2 .

[0179] A current sense resistor **2015** that is in series with the load **2010** is coupled to a switch network **2020** and to sampling capacitors C_1 and C_2 . The switch network sequentially samples the potential across the current sense resistor **2015** at an interval controlled by the sampling control clock **2025**. The current sense resistor may be a specific discrete resistor that is added to the circuit, or it may be a resistance that is intrinsic to the circuit.

[0180] At the beginning of a sampling cycle capacitor C_1 may be switched by the switch network **2020** to a parallel connection with the current sense resistor **2015** for a short period of time (e.g., <10 nanoseconds) that allows C_1 to track the potential across the current sense resistor **2015**. C_1 is then disconnected from the current sense resistor **2015** by the switch network **2020**. The sampling process may then be repeated for C_2 . After voltage samples V_1 and V_2 have been acquired respectively on C_1 and C_2 , the switch network **2020** subsequently couples C_1 and C_2 to comparator **2030**.

[0181] When charging the capacitance associated with the load **2010** by a fixed voltage in the absence of redox reactions, the current through the current sense resistor **2015** will decrease over time. Since V_2 is acquired after V_1 , it will normally be less than V_2 . However, the onset of a redox reaction may produce an increase in current that will result in V_2 being greater than V_1 . When this happens, the comparator output changes state, causing the control logic to turn off the driver **2045** which in turn causes the power switch **2005** to shut off. Alternatively, the power switch may reduce the current to a preselected value for a period prior to shutting off.

[0182] RC measurement circuit shown in FIG. **19** may be used to establish process pulse waveform parameters prior to applying the process pulse waveform to an electrolytic cell. In contrast, the redox detection circuit shown in FIG. **20** may be used to provide realtime control of a process pulse waveform on a pulse-by-pulse basis. The RC measurement circuit shown in FIG. **19** and the redox detection circuit of FIG. **20**

may each be used in conjunction with the circuits shown in FIG. 11, FIG. 12A, and FIG. 12B.

[0183] FIG. 21A shows an electrical schematic for an embodiment of an electrolytic isotope separation system 2100. A transmission line duct 2115 includes two basic units, a bus transmission line segment 2115a and a transmission line electrode unit 2115b. The bus transmission line segment 2115a is represented by series inductance L_{it} , series resistance R_{it} , and shunt capacitance C_{it} . The transmission line electrode unit 2115b is represented by series inductance L_{series} , series resistance R_{series} , combined with an electrolytic cell represented by a combination of double layer capacitance C_{dl} , inductance L_{shunt} , and electrolyte resistance R_{el} in parallel with a charge transfer resistance R_{ct} . For simplicity, dielectric and magnetic loss elements are not shown, although they may be significant at high frequencies.

[0184] A pulse power supply 2105, a radio frequency (RF) power supply 2110 and shunt 2120 are each coupled by a pair of switches 2106 to the transmission line duct 2115. Two switches generally provide better isolation between the switched components, and reduce parasitic elements (e.g., capacitance) seen by an active component. Alternatively, a single switch may be used with the other switch being replaced by a connection.

[0185] The pulse power supply 2105 provides current for charging/discharging the double layer capacitance C_{dl} and/or carrying out redox reactions. For example, the circuit shown in FIG. 11, or a derivative thereof, may serve as the pulse power supply 2105. The pulse power supply 2105 is generally switched in to the transmission line duct 2115 when the RF power supply 2110 and shunt 2120 are switched out, although a DC bias may be combined with an RF signal. In general, it is desirable that the total inductance for the transmission line structure be less than one microhenry and that the RC time constant of the electrolytic cell be less than one millisecond. These electrical constraints translate to close conductor and electrode spacings and a compact system size when realized in a physical embodiment. When a high frequency magnetic excitation is to be applied, the electrical requirements become more stringent.

[0186] The RF power supply 2110 may be used to provide a high frequency current at one or more frequencies. The RF power supply 2110 may include one or more oscillators, which may be either tunable or fixed frequency. The RF power supply 2110 is typically used in conjunction with the shunt 2120. The shunt 2120 provides a switchable path that reduces the voltage developed across the electrolytic cell (R_{ct} , C_{dl} , L_{shunt} , and R_{el}), while providing a current through the transmission line conductors. The shunt 2120 may act as a short circuit in a lumped circuit, or may provide a matched termination to minimize reflections in a distributed circuit.

[0187] Depending upon the physical dimensions associated with the transmission line duct 2115, the nature of the dielectric used in construction, and the operating frequency of the RF power supply 2110, the transmission line duct 2115 may be treated as either a lumped circuit or a distributed circuit. In general, it is desirable that size of the electrolytic isotope separation circuit 2100 be chosen so that it may be treated as a lumped circuit; however, at high frequencies (e.g., above about 100 MHz), the difficulty associated with physical miniaturization must be balanced with the complexity of dealing with a distributed circuit.

[0188] An array of small circuits that can be treated individually as lumped circuits is preferred to a single large

system that has a dimension on the order of the excitation wavelength. Nuclear magnetic resonance (NMR) frequencies in low static magnetic fields are generally below 100 MHz, whereas electron paramagnetic resonance (EPR) frequencies may be orders of magnitude higher (e.g., greater than 10 GHz). Due to skin depth effects, a large system will have resistive losses associated with conductor lengths that cannot be simply offset by a proportional increase in conductor cross-section. In a preferred embodiment, the length of the electrode/electrolyte interface, measured in the direction of the current flow, is less than $1/100$ of the wavelength of the magnetic excitation frequency.

[0189] The combination of the RF power supply 2110 and transmission line duct 2115 may be a resonant structure with capacitance being largely provided by the RF power supply 2110, and inductance being largely provided by the transmission line duct 2115 series inductance L_{series} . The resonant frequency of the structure may be tuned to a frequency for producing a microwave-induced magnetic isotope effect (MIMIE) in species present in the electrolyte within the transmission line electrode unit 2115b.

[0190] FIG. 21B shows an electrical schematic for an embodiment of an electrolytic isotope separation system 2101 with a pulsed excitation source 2130 coupled to a transmission line duct 2125. The pulsed excitation source includes a charging circuit 2132 coupled to a pulse capacitor C_{pulse} by a switch 2106d. C_{pulse} is coupled to a switch diode D_{sw} by a switch 2106c. Resonant capacitor C_{res} and D_{sw} are coupled to the transmission line duct 2125 by switch 2106e.

[0191] In the following description of an operational embodiment it is assumed that switches 2106(a, b, c, d) are initially open. Excitation is enabled by closing switches 2106a and 2106b so that shunt 2120 effectively shorts the end of the transmission line duct 2125. Switch 2106d is closed to charge C_{pulse} . Once C_{pulse} is charged, switch 2106d may be opened. Switch 2106c is closed to charge C_{res} . Switch 2106e may then be closed to connect pulsed excitation source 2130 to the transmission line duct 2125. Upon the closure of switch 2106e, the energy stored in C_{res} will oscillate between C_{res} and the inductance L_{series} of transmission line 2125. R_{series} and other lossy elements will damp the oscillation, which may be regenerated by further discharges from C_{pulse} . It should be noted that a single capacitor may be used for single shot excitation.

[0192] In one embodiment, switch 2106e is kept in a closed state while switch 2106c is operated to produce a sequence of resonance regeneration pulses. Each pulse produced by switch 2106c produces a damped resonant response that decays after a number of cycles at the resonant frequency. For example, transmission line duct system 2101 may be excited at a resonant frequency of 80 MHz and switch 2106c may be operated at a frequency of 10 MHz. In the process of excitation, a precise amplitude may not be critical, but it may be desirable to maintain the current above a threshold value required for a desired magnetic field intensity within the electrolyte gap. The number of resonant cycles between pulses applied by switch 2106c may be determined by the threshold current value and the amount of energy delivered in each pulse. Thus, an excitation frequency that is considerably higher than the power switching frequency may be obtained.

[0193] Selection of the value for C_{res} may be done by characterizing the transmission line 2125 and then selecting the value of C_{res} that corresponds to a desired resonant frequency. The characteristics of electrolytic cell 2135 may vary with

frequency, particularly at high frequencies (e.g., the Debye-Falkenhagen effect). Thus, the criteria for selection of solvents and/or electrolytes may extend beyond electrochemical concerns and may involve solvent and/or ionic species behavior at RF and microwave frequencies.

[0194] Typically, the required excitation voltage will rise with frequency due to the increasing inductive reactance. However, the increase in applied frequency will offset the enhanced charging of C_{dl} due to the increased voltage since the charging time will be reduced. It is generally desired that redox reactions in the electrolytic cell be avoidable during RF or microwave excitation. The area of the electrode/electrolyte interface may be selected to provide a desired C_{dl} . R_{el} may also be tailored to provide an RC time constant that allows RF excitation to be achieved while minimizing unwanted redox reactions in the electrolytic cell during excitation. An intentional DC bias may be applied to induce redox reactions during excitation.

[0195] FIG. 22A shows a perspective view of an embodiment of a parallel plate transmission line duct 2200 with shunt switches 2220. A top plate 2235 has an aperture 2230 and electrolyte ports 2225. Shunt 2215 is coupled to a top conductor 2210 by switches 2220. It is preferable that switches 2220 have low inductance and low resistance in the on state. Top conductor 2210 is disposed between a dielectric 2205 and top plate 2235.

[0196] FIG. 22B shows an exploded view 2201 of the parallel plate transmission line duct 2200 shown in FIG. 27A. A bottom conductor 2240 is separated from the top conductor 2210 by the dielectric 2205. It is preferable that the dielectric 2205 be a material with a low dielectric constant and low dielectric loss, particularly at high frequencies. Fluorocarbon polymers and porous materials may be used. A portion of the dielectric may be a material with a high magnetic permeability that serves to modify a magnetic field in the gap between the top conductor 2210 and the bottom conductor 2240.

[0197] Preferred materials for the top conductor 2210 and bottom conductor 2240 are copper and silver, particularly at high frequencies where the skin depth is small. The top conductor 2210 and bottom conductor 2240 may have portions that are coated to provide compatibility with an electrolyte or liquid metal. For example, the top conductor 2210 may have a platinum coating. It is generally desirable to maintain thin (e.g., less than one micron) coatings of uniform thickness with abrupt transitions to the base metal, particularly at higher frequencies. A liquid metal stabilizer 2245 and an electrode chamber 2250 provide containment for a liquid metal electrode. In an alternative embodiment, the liquid metal stabilizer is a narrow slit with a length that is at least ten times greater than its width. The electrode chamber 2250 may have one or more ports 2255.

[0198] Ideally, most of the magnetic flux produced by excitation current flowing in the parallel plate transmission line duct 2200 would pass through the electrolyte chamber 2206; however, for a structure with a uniform magnetic permeability the magnetic field will be distributed over a region of space that will be significantly larger than the volume of the electrolyte chamber 2206. Thus, it may be desirable to introduce elements that can shape the magnetic field and increase the magnetic flux that is produced by a given excitation current.

[0199] A magnetic field enhancer 2260 intensifies the magnetic field in the electrolyte chamber 2250 that is produced by the RF current flowing through the parallel plate transmission line duct 2200. The magnetic field enhancer is fabricated

from a material that has a relative permeability greater than one. In general, structures with a high initial permeability and a low saturation inductance are preferred. In a particular embodiment the magnetic field enhancer 2260 is saturated at current levels that exceed the desired operating excitation current amplitude by 10% or more.

[0200] In order for the parallel plate transmission line duct 2200 to provide a sufficiently strong RF magnetic field within the electrolyte chamber 2206, a certain amount of inductance is required. However, too much inductance in the electrolytic current path may degrade the electrolytic pulse waveform. Since the peak electrolytic current may be much larger than the current used to produce the RF magnetic field, a low saturation inductance in the magnetic field enhancer 2260 minimizes the impact on the electrolytic pulse waveform that is applied. Application and synchronization of the electrolytic pulse and magnetic excitation may be controlled by components similar to those disclosed with respect to FIG. 7A.

[0201] A magnetic field enhancer 2260 may be disposed between the upper conductor 2210 and the lower conductor 2240. Since a magnetic field enhancer 2260 that is disposed between the upper conductor 2210 and the lower conductor 2240 will have a greater impact on the capacitance of the parallel plate transmission line duct 2200, a material with a lower dielectric constant may be used. Although the dielectric constants of the materials of construction are not critical with respect to the electrolytic pulse, it can make the difference as to whether the parallel plate transmission line duct 2200 may be treated as a lumped circuit or a distributed circuit, as the excitation wavelength in the transmission line duct 2515 will decrease with increasing dielectric constant of the dielectric 2205.

[0202] In a particular embodiment, a portion of a parallel plate transmission line duct 2200 that includes a magnetic field enhancer 2260 and an electrolyte chamber 2206, forms a magnetic circuit in which more than 75% of the shortest magnetic flux path length lies within the magnetic field enhancer 2260 and less than 25% of the magnetic flux path lies within the electrolyte chamber 2206.

[0203] In one embodiment the magnetic field enhancer 2260 is fabricated from a homogeneous soft ferrite. In another embodiments, thin film laminate and/or composite structures may be used. More than one magnetic field enhancer 2260 may be used to enhance and/or shape the RF magnetic field. Since the RF current path differs from the electrolytic current path, the magnetic field enhancer may not be placed to simply maximize the RF magnetic field intensity, but may be placed to optimize the tradeoff between the increase in the RF magnetic field and the degradation of the electrolytic pulse.

[0204] A window insert 2211 provides for the transmission of electromagnetic radiation to the electrolyte chamber 2206. The window insert is preferably a high transmittance material that is chemically inert with respect to the electrolyte that is used. The window insert may have a transparent conductive coating such as indium tin oxide, or it may have a metal pattern disposed on the surface to form an electrode. In a preferred embodiment, a metal pattern having parallel metal traces with a width of less than 20 microns is used. Diffraction of incident electromagnetic radiation should be taken into account when a metal pattern is used, particularly for monochromatic sources.

[0205] FIG. 22C shows a section view 2202 of the parallel plate transmission line duct 2200 shown in FIG. 22A. The cavity in the electrode chamber 2250 allows a contained

liquid metal to electrically couple the bottom conductor **2240** to the shunt **2215**. Thus, the electrode chamber **2250** may be either a dielectric or conductor. The liquid metal stabilizer **2245** separates the electrode chamber from the electrolyte chamber **2206**. Access to the electrolyte chamber **2206** is obtained through the electrolyte ports **2225**. An electrolyte circulation system may be used to remove heat produced by the electrolytic current and excitation energy.

[0206] FIG. 22D shows a section view of an embodiment of a parallel plate transmission line duct **2203** similar to that shown in FIG. 22C, except that a solid electrode **2241** replaces the liquid metal electrode assembly and is coupled directly to the shunt switches **2220**. Solid electrodes are preferred for electrolytic processes involving oxidation or partial reduction of species, particularly in a low conductivity electrolyte where a narrow gap is desired. A magnetic field enhancer **2261** is coupled to the solid electrode **2241**. Although the separation between electrodes **2212** and **2241** is shown as uniform, the separation may be varied along the length of the transmission line duct **2203**. For example, the separation may be greater across the electrolyte gap **2206** and smaller across the dielectric **2205**, or vice versa.

[0207] FIG. 23 shows a perspective view of a parallel plate transmission line duct system **2300**. A parallel plate transmission line duct **2305** is magnetically coupled to a single turn solenoid **2306**. An aperture **2310** allows the electrolytic cell portion **2325** of the transmission line duct **2305** to reside within the core of the single turn solenoid **2306**. The top conductor **2315** and bottom conductor **2320** extend beyond the core of the single turn solenoid **2306**. The single turn solenoid **2306** may be used to provide a magnetic field for RF excitation of species within the electrolytic cell **2325** and provides an alternative to the self-excited structure shown in FIG. 22A. Supporting dielectric structures and magnetic materials similar to those disclosed elsewhere within this application may be used to provide mechanical support and magnetic field modification, and are not shown in FIG. 23. In general, an external magnetic field source other than a single turn solenoid may be used and may be configured in other orientations.

[0208] FIG. 24 shows a flow diagram **2400** for an embodiment of an isotope separation process. At step **2405**, an exclusion pulse is applied to an electrolytic cell. The exclusion pulse establishes a desired concentration profile for cationic and anionic species within the interphase at an electrode of the electrolytic cell. The potential applied to an electrode is the same as the charge of the species of interest at that electrode. For example, if a cationic species such as a metal cation (e.g., U^{4+} or Cu^{2+}) or metal containing complex (e.g., UO_2^{2+}) is the species of interest, a positive potential is applied. The applied positive potential decreases the concentration of the species of interest within the interphase. Similarly, if an anionic species such as a metal complex (e.g., UCl_6^{2-} or $UO_2Cl_4^{2-}$) is the species of interest, a negative potential is applied.

[0209] The exclusion pulse creates a depletion region through which the species of interest may subsequently be transported to the electrode surface under the influence of an applied potential of opposite potential to that of the exclusion pulse. The products of reactions involving the species of interest may also be subsequently transported to the electrode surface under the influence of an applied potential. The increased separation between the electrode surface and the species of interest provides a greater distance over which

mass-dependent transport processes (e.g., electromigration and diffusion) may provide isotope separation.

[0210] Conventional centrifuge and diffusion techniques for isotope separation typically rely on the gaseous state and thus have a relatively limited number of compounds that can be used as a working material. In contrast, there are an enormous number of anionic and cationic species that can be prepared using a wide variety of solvents and solutes. Water, aprotic solvents, molten halides, and room temperature ionic liquids are examples of solvents that may be used. Given the wide variety of organic and inorganic liquids, and supercritical fluids that are available for use, a mixture of isotopes of any of the following elements may be prepared as a dissolved ionic species, organometallic compound, or a soluble complex for use in embodiments of the present invention: Li, B, C, Mg, Si, K, Ca, Ti, V, Cr, Fe, Ni, Cu, Zn, Ga, Ge, Se, Rb, Sr, Zr, Mo, Ru, Pd, Ag, Cd, In, Sn, Sb, Te, Ba, La, Ce, Nd, Sm, Eu, Gd, Dy, Er, Yb, Lu, Hf, Ta, W, Re, Os, Ir, Pt, Hg, Tl, Pb, Bi, Po, Th, U, Np, Pu, Am, and Cm.

[0211] Room temperature ionic liquids (RTILs) are of particular interest since there are many possible compounds that can be prepared. The selection of cation and anion for a RTIL can take into account the properties desired in an ionic species (e.g., a transition metal or actinide complex). For example, quaternary ammonium salts of the bis(trifluoromethanesulfonyl)imide anion ($N(SO_2CF_3)_2$ (i.e., $-NTf_2$) have been shown to be useful vehicles for redox reactions involving uranium and uranium complexes. A room temperature ionic liquid may be used in combination with salts (e.g., chlorides) that provide additional complexing agents or ligands.

[0212] In one embodiment, the exclusion pulse appears across the electrode surfaces as a fixed voltage square wave with a rise time of less than one microsecond and a fall time of less than microsecond, and the applied voltage is a voltage at which no redox reactions occur that involve species other than impurities. In another embodiment, the exclusion pulse appears across the electrode surfaces as a fixed voltage square wave with a rise time of less than 500 nanoseconds and a fall time of less than 500 nanoseconds, and the applied voltage is a voltage at which no redox reactions occur that involve species other than impurities. Further, in each of the aforementioned embodiments the RC time constant of the electrolytic cell associated with the electrodes is greater than 10 microseconds and less than 1000 microseconds.

[0213] Cationic or anionic species may be excluded at an electrode. For example, a UO_2^{2+} cationic complex may be excluded at an electrode by applying a positive potential prior to reversing the electrode polarity and carrying out a reduction of the UO_2^{2+} cationic complex. Similarly an anionic trivalent actinide (e.g., U^{3+}) complex may be excluded at an electrode by applying a negative potential prior to reversing the potential and carrying out an oxidation of the anionic trivalent actinide complex.

[0214] At step **2410**, the species of interest is dissociated by the application of an energy pulse. The pulse of energy may be electromagnetic radiation (e.g., with a wavelength between 0.2 microns and 20 microns). The species of interest may be an electrically neutral species such as an organometallic compound, a solvated ion, or a charged complex (e.g., transition metal or actinide complex). The dissociation of the species of interest may result in a radical pair or a radical-ion pair, which may be spin-correlated. The radical pair or radical-ion pair may be a triplet pair or may be a singlet pair.

[0215] For gaseous atoms or free ions, distinct differences in optical absorption may exist between isotopes. However, in solutions the differences are less distinct due to the effects of the solution environment. A monochromatic or narrow-band light source may be used to produce a small preferential dissociation for a species containing a particular isotope. This preferential dissociation may contribute to an overall isotope separation process that employs the mass isotope effect or the magnetic isotope effect.

[0216] The use of liquid electrolytes also allows for exploitation of the field shift effect in which a heavy isotope has a preference for the chemical species with the smaller number of s electrons in the bonding or valence orbital. With the proper selection of photonic and magnetic excitations and reactant species, the magnetic isotope effect and the field shift effect can be exercised in concert to provide an enhanced overall isotope separation.

[0217] An anionic complex may be dissociated to produce an anion-cation pair or a neutral-anion pair. Similarly, a cationic complex may be dissociated to produce an anion-cation pair or a neutral-cation pair. A pair produced by dissociation may also be further dissociated to form another pair through multi-photon absorption. Pair formation may involve electron transfer with other adjacent species (e.g., photoreduction).

[0218] At step **2415**, magnetic excitation is applied. The excitation may be applied by a DC magnetic field, an alternating magnetic field, or a combination of a DC magnetic field and an alternating magnetic field. A DC magnetic field may be applied to modify the spin evolution of species formed in step **2410**. The Δg mechanism (ΔgM), hyperfine coupling mechanism (HFCM), and the level crossing mechanism (LCM) are examples of mechanisms that can be modified by a DC magnetic field to control differential spin conversion of isotope containing radical pairs or radical-ion pairs. An alternating magnetic field may also be used to induce differential level transitions in the electron and nuclear spins of isotope containing species. The alternating magnetic field may have a frequency in the range of 100 kHz to 100 GHz. Although the energy differences involved in spin conversion may be small in comparison to the thermodynamic energies associated with chemical reactions, they may have a significant impact on chemical reaction rates.

[0219] Excitation by an alternating magnetic field or a combination of an alternating magnetic field and a DC magnetic field may be used to alter the relative recombination or reaction rates of magnetic and nonmagnetic isotope containing pairs. The alteration of recombination or reaction rates may include accelerating or retarding a reaction rate. The excitation may be used to produce a transient population of cationic or anionic species having an enhanced concentration of a magnetic or nonmagnetic isotope. Although not as great as the differences between magnetic and nonmagnetic isotopes, the difference in magnetic moment between magnetic isotopes of the same element (e.g., ^{63}Cu and ^{65}Cu) may also be used as a basis for transient fractionation through magnetic excitation.

[0220] Transient fractionation produced by managed differences in nuclear spin and magnetic moment may be used to produce an isotopically enhanced population near the surface of an electrode that may be converted to stable species through redox reactions at the electrode surface. In addition to the enhancement of the recombination and/or reaction of magnetic isotopic species to produce such a population, magnetic excitation (e.g., spin inversion) may be used to provide

a relative enhancement of recombination and/or reaction of nonmagnetic isotopic species with respect to magnetic isotopic species to create the population.

[0221] Reversible photoreduction of uranyl (UO_2^{2+}) to uranoyl (UO_2^+) in the presence of an appropriate electron donor provides a method for transient fractionation that relies on magnetically enhanced reoxidation of $^{235}\text{UO}_2^+$ to $^{235}\text{UO}_2^{2+}$ to provide a ^{235}U enhanced population of UO_2^{2+} that may be attracted to an electrode with a greater velocity than the ^{238}U enriched UO_2^+ . As with other photolytic transient fractionation processes that rely on differential recombination to reproduce a starting species, it is desirable to have a high quantum yield for the initial reaction.

[0222] At step **2420**, a potential is applied to the electrolytic cell to extract cationic or anionic species. The population of cationic or anionic species attracted to the electrode may or may not have been produced by step **2405** and/or step **2410**. In the absence of step **2405** and **2410**, the mass isotope effect will be the primary effect in providing isotope separation. As the ionic species migrate toward the electrode, the lighter isotope will do so with a greater velocity, causing the initial contact population on the electrode surface to be enriched in the lighter isotope.

[0223] The applied potential waveform may correspond to the (V_1, t_1) and/or (V_2, t_2) segments shown in FIG. 9. In one embodiment, the attraction pulse results in a current peak of at least 20 amperes and appears across the electrode surfaces as one or more fixed voltage square waves, with each having a rise time of less than 500 nanoseconds and a fall time of less than 500 nanoseconds. In another embodiment, the attraction pulse results in a current peak of at least 50 amperes and is applied across the electrode surfaces as one or more fixed voltage square waves, with each having a rise time of less than 100 nanoseconds and a fall time of less than 100 nanoseconds. Further, in each of the aforementioned embodiments the RC time constant of the electrolytic cell associated with the electrodes is greater than 10 microseconds and less than 1000 microseconds.

[0224] Transient isotope fractionation provided by step **2405** and/or step **2410** may be used to produce a population of cation/neutral pairs from a cationic complex, with the difference in mass between the cationic complex and the cation pair component being considerably greater than the isotope mass difference. For example, accelerated recombination of a ^{235}U containing pair will produce a population of lighter unrecombined cations that is enriched in ^{238}U . Since most neutral species (e.g., solvent molecules) will have a mass that is considerably greater than the 3 atomic mass unit difference between ^{235}U and ^{238}U , the total isotope fractionation at the electrode surface will be a combination of initial population isotope fractionation through the magnetic isotope effect combined with an enhanced mass isotope effect during migration. Although all cationic species will respond to the electrode potential, the ^{238}U containing species will be greater in number and faster than the ^{235}U containing species.

[0225] In another embodiment, transient isotope fractionation between magnetic and nonmagnetic isotopes is provided by step **2405** and/or step **2410** to produce a population of cation/anion pairs from an anionic complex. Due to enhanced recombination and/or reaction of magnetic cations (e.g., ^{235}U containing cations) to form anionic complexes, the unrecombined cation population will be rich in nonmagnetic (e.g., ^{238}U containing cations). Under the influence of a negative potential at the electrode surface, the ^{235}U rich population

of anionic complexes will tend to be excluded from the electrode surface as the ^{238}U rich cation complexes are attracted to the electrode surface.

[0226] Upon application of an extraction pulse, both diffusion and migration within the applied electric field may drive mass transport to the electrode. It is desirable that the transport time to the electrode surface be shorter than the lifetime of transient species of interest. The mean distance to the electrode may be decreased by increasing the concentration of ionic species, and the transport velocity may be increased by reducing the electrolyte viscosity. The magnitude of the exclusion potential applied in step 2405 may be reduced to decrease the mean distance to the electrode.

[0227] At step 2425, charge transfer between the electrode and species attracted to the electrode surface during step 2420 occurs and an oxidation or a reduction is carried out. The oxidation or reduction reaction may be partial or complete. For example, a Cu^{2+} cation may be reduced to Cu^+ or it may be reduced to Cu metal. Reduction may be carried out at a solid or liquid electrode surface. The charge involved in the reaction may be provided by the attraction pulse applied in step 2420.

[0228] FIG. 25A shows a timing diagram 2500 for an embodiment of an isotope separation process with non-overlapping pulses and a simple extraction pulse. A photolytic energy waveform PH_{cell} is applied for a period t_1 . Subsequent to the application of waveform PH_{cell} , a magnetic excitation waveform ME_{cell} is applied with a period t_2 . Following the application of waveform ME_{cell} , an extraction pulse V_{cell} is applied for a period t_3 . In an embodiment, t_1 is less than one microsecond, t_2 is less than one microsecond, and t_3 is less than 10 microseconds. Non-overlapping pulses are generally preferred for a sequence in which a precursor is dissociated then subjected to differentially enhanced recombination to produce isotope fractionation, then extracted to the electrode for a redox reaction.

[0229] FIG. 25B shows a timing diagram 2501 for an embodiment of an isotope separation process with non-overlapping pulses similar to that shown in FIG. 30A, except that the simple extraction pulse with a period t_3 has been replaced with a complex extraction pulse similar to that shown in FIG. 9, having period segments t_3 , t_4 , and t_5 . The complex pulse is preferred for electrolytic cells with a large RC time constant (e.g., greater than 10 microseconds), and an extraction species with a short lifetime (e.g., less than 10 microseconds).

[0230] FIG. 25C shows a timing diagram 2502 for an embodiment of an isotope separation process with a magnetic excitation ME_{cell} pulse that overlaps an extraction V_{cell} pulse. Overlapping excitation and extraction pulses may be used in a process in which magnetic excitation is used to inhibit recombination or reaction (e.g., spin locking), and thus maintain fractionation during extraction.

[0231] FIG. 25D shows a timing diagram 2503 for an embodiment of an isotope separation process with overlapping photolytic PH_{cell} and extraction V_{cell} pulses. Overlapping photolytic and extraction process may be used in a process in which a degree of fractionation is achieved through photolysis and augmented by a mass isotope effect during transport to the electrode.

[0232] FIG. 26 shows an electrical schematic diagram 2600 for an embodiment of an electrolytic cell with a magnetic excitation source 2610 coupled to a working electrode 2625 by a transmission line 2615. Magnetic excitation source 2610 may be configured to provide a single pulse or a pulse train.

When configured to provide a single pulse, optional capacitance C_{tune} may be used to establish a resonant frequency for the current flowing through magnetic excitation source 2610, transmission line 2615, and working electrode 2625. Transmission line 2615 includes a $C1_{\text{shunt}}$ that is effectively in parallel with C_{tune} , as well as series inductances $L1a_{\text{series}}$ and $L1b_{\text{series}}$, and series resistances $R1a_{\text{series}}$ and $R1b_{\text{series}}$. Working electrode 2625 includes series inductances $L3a_{\text{series}}$ and $L3b_{\text{series}}$ and series resistances $R3a_{\text{series}}$ and $R3b_{\text{series}}$. It is preferable that the sum of $L3a_{\text{series}}$ and $L3b_{\text{series}}$ is greater than the sum of $L1a_{\text{series}}$ and $L1b_{\text{series}}$, since the magnetic excitation is provide principally by the magnetic field associated with $L3a_{\text{series}}$ and $L3b_{\text{series}}$.

[0233] A redox power supply 2605 is coupled to working electrode 2625 and alternate electrode 2635 by a transmission line 2620. Redox power supply 2605 may be similar to the power supplies shown in FIG. 10 and FIG. 11. Working electrode 2625 and alternate electrode 2635 correspond to an electrode assembly, and are coupled by an electrolyte 2630 characterized by a double layer capacitance C_{DL} and a series resistance R_{EL} . Alternate electrode 2635 includes a series inductance $L4a_{\text{series}}$ and a series resistance $R4a_{\text{series}}$. Transmission line 2620 includes a $C2_{\text{shunt}}$, series inductances $L2a_{\text{series}}$ and $L2b_{\text{series}}$, and series resistances $R2a_{\text{series}}$ and $R2b_{\text{series}}$.

[0234] The impedance of redox power supply 2605, electrolyte 2630, and $C2_{\text{shunt}}$ is such that the current produced by magnetic excitation source 2610 is essentially limited to a single loop through working electrode 2625 without flowing through the electrolyte 2630 and the alternate electrode 2635. This configuration reduces the damping effect of R_{EL} during resonant excitation.

[0235] In an embodiment, transmission line 2615 has a capacitance in excess of 400 pF per meter and an inductance of less than 50 nH per meter. Similarly, it may also be desirable that transmission line 2115a of FIG. 21A have a capacitance in excess of 400 pF per meter and an inductance of less than 50 nH per meter. The requirement for chip capacitors may be reduced by taking advantage of the inherent capacitance of the transmission line structure in resonant applications.

[0236] FIG. 27 shows a perspective view 2700 of an embodiment of an electrolytic cell 2705 with a magnetic excitation transmission line 2710 that corresponds with the schematic shown in FIG. 26. In this embodiment, the electrolytic cell 2705 is electrically connected to the magnetic excitation transmission line 2710. In other embodiments, the electrolytic cell 2705 may be isolated from the magnetic excitation transmission line 2710 by a thin dielectric. The use of a common working electrode that is part of both the electrolytic cell 2705 and the magnetic excitation transmission line 2710 generally provides for a maximum magnetic field intensity at the surface of the common working electrode. However, at high frequencies skin depth and proximity effect considerations may result in a preference for different conductive materials for the magnetic transmission line 2710 and the working electrode in the electrolytic cell 2705.

[0237] FIG. 28 shows an exploded view of the electrolytic cell 2705 of FIG. 27. A first plate conductor 2810 and a second plate conductor are separated by a dielectric 2820. The first plate conductor 2810 connects to an alternate electrode 2815 that is supported by a frame 2825. The second plate conductor 2830 is connected to a working electrode 2840 by a contact 2835. The working electrode 2840, frame

2825, and cover **2805** provide an enclosure for a volume of electrolyte. Cover **2805** may be transparent so that the enclosure may be illuminated. In other embodiments the discrete working electrode **2840** may be omitted, with the frame **2825** and contact **2835** being connected to the surface of the excitation transmission line **2710** in FIG. 27. Electrode **2815** may be a metal conductor with apertures for light transmission, or it may be a transparent conductive coating.

[0238] FIG. 29 shows an exploded view of the magnetic excitation transmission line **2710** of FIG. 27. A top conductor **2905** and a bottom conductor **2925** are separated from a center conductor **2915** by insulators **2910** and **2920**, respectively. When assembled, the right hand ends **2935** of conductors **2905**, **2915**, and **2925** are in electrical contact with each other, as shown in FIG. 27. Top conductor **2905** and bottom conductor **2925** each have an aperture **2940** that allows for a locally more intense magnetic field near the narrowed portion **2945** of center conductor **2915**. An optional ferrite mass **2930** may be placed in close proximity to the narrowed portion **2945** to enhance the magnetic field. In other embodiments the top conductor **2905** or bottom conductor **2925** may be omitted.

[0239] FIG. 30 shows an embodiment of a photolytic transmission line duct **3000**. A photolytic duct segment **3005** is coupled to a magnetic excitation duct segment **3020**. An electromagnetic radiation source **3015** illuminates a working fluid **3035** (e.g., gas or liquid) that flows from the photolytic duct segment **3005** through the magnetic excitation duct segment **3020**. In this embodiment duct **3010** has a greater cross-section than duct **3025**, producing a greater fluid velocity in the magnetic excitation duct segment **3020**. A magnetic excitation transmission line **3030** provides an alternating magnetic field within duct **3025**. Species within the working fluid **3035** may be photoexcited within the photolytic duct segment **3005** and subsequently subjected to spin conversion or spin locking within the magnetic excitation duct segment **3020**. The photoexcitation may be isotope selective, or may be achieved with a broadband source such as a flash lamp. In other embodiments the magnetic excitation transmission line **3030** may be replaced by a single turn solenoid that wraps around the duct **3025**.

[0240] FIG. 31 shows an embodiment of a cascade isotope separation system **3100**. A photolytic transmission line duct **3105** similar to that shown in FIG. 32 is coupled to a mass effect isotope separation device **3115**. Mass isotope separation device may be an aerodynamic separation device, a vortex tube, or other device. The input stream **3110** contains isotopologues that are subjected photolysis and magnetic excitation to produce product streams **3120** and **3125** that are mass differentiated on the basis of their isotopic composition.

[0241] FIG. 32 shows an embodiment of a serial transmission line duct reactor **3200**. An electrolyte **3220** flows from a photolytic duct segment **3205**, through a magnetic excitation duct segment **3225**, and to a redox duct segment **3235**. Photolysis of species within the electrolyte **3220** is provided by photon source **3215**. Magnetic excitation is provided by magnetic excitation source **3230**. Duct **3210** has an initial dimension of W_1 in the photolytic duct segment **3205** that narrows to W_2 within the magnetic excitation **3225** and redox segment **3235**. In the serial transmission line duct reactor **3200**, the redox and magnetic excitation functions are spatially separated, and thus cannot be used for magnetically excited species that have very short lifetimes. However, for metastable magnetically excited species with a lifetime greater than the

transit time from the magnetic excitation duct segment **3225** to the redox duct segment **3235**, anode **3240** and cathode **3245** can be used at a selected voltage without requiring pulses. Although the range of usable species is limited, there is a greater efficiency since the double-layer capacitance does not have to be continuously charged and discharged.

[0242] FIG. 33 shows an embodiment of a magnetic excitation transmission line **3300**. A center conductor **3305** is coupled to a return conductor **3310** that is orthogonal to the center conductor **3305**. A ferrite body **3315** provides enhancement of the magnetic field at the surface of the center conductor **3305**. Although structure of magnetic transmission line **3300** is less compact than the magnet excitation transmission line **2710** shown in FIG. 27, at high frequencies with significant skin depth and proximity effects it may provide a more desirable magnetic excitation pattern near the surface of center conductor **3305**. FIG. 34 shows an embodiment of a coaxially coupled magnetic excitation transmission line **3400**. A coaxial outer conductor **3410** is coupled to an orthogonal return conductor **3420** and a coaxial inner conductor is coupled to a center conductor **3415**. A ferrite body **3425** provides enhancement of the magnetic field at the surface of the center conductor **3415**.

[0243] FIG. 35 shows a cross-section view of an embedded magnetic excitation transmission line reactor **3500**. A substrate **3505** supports a ground plane **3515** and electrodes **3510**, **3520**, and **3525** that are in contact with a liquid electrolyte **3530**. Liquid electrolyte may be essentially static, or it may be flowing parallel to the surface of the substrate **3505**. A transparent cover **3535** may be used to permit photoexcitation and limit the thickness of the electrolyte **3530**.

[0244] For configurations in which the electrolyte **3530** is flowing, a magnetic excitation current is established between electrode **3510** and ground plane **3515** and a redox potential is established between electrode **3520** and electrode **3525**. For configurations in which the electrolyte **3530** is essentially static, electrode **3510** is omitted and the magnetic excitation current is applied between the ground plane **3515** and either electrode **3520** or electrode **3525**. Substrate **3505** may be a dielectric (e.g., sapphire) or a semiconductor (e.g., silicon). For semiconductor substrates, the electrodes **3510**, **3520**, **3525**, and ground plane **3505** are isolated from the substrate **3505** by thin dielectric films.

[0245] While the invention has been described in detail with reference to preferred embodiments thereof, it will be apparent to one skilled in the art that various changes can be made, and equivalents employed, without departing from the scope of the invention. Various embodiments of power supplies, transmission line structures, fluid circuits, and electrode assemblies have been disclosed using a variety of components. Within the scope of the invention, combinations of the aforementioned disclosed components other than those combinations explicitly disclosed may be used in a system for isotope selective chemical reactions.

What is claimed:

1. A system for spin modification of an electrolytic process, said system comprising:

an electrolyte containing at least two spin-bearing species;
at least one first electrode in contact with said electrolyte;
at least one second electrode in contact with said electrolyte;

An alternating current source isolated from said second electrode and coupled to said first electrode for providing an alternating current through said first electrode,

wherein said current produces an alternating magnetic field in a portion of electrolyte adjacent to said first electrode, thereby modifying the spin of at least one species in said portion of electrolyte adjacent to said first electrode; and,

a current source coupled to said first electrode and to said second electrode for electrolyzing at least a portion of at least one of said two spin-bearing species at said first electrode.

2. The system of claim 1, wherein said alternating magnetic field comprises at least one frequency between 100 kHz and 100 GHz.

3. The system of claim 1, wherein said alternating magnetic field comprises more than one frequency.

4. The system of claim 1, further comprising a magnet for providing a DC magnetic field, wherein said magnetic field manipulates the Zeeman interaction of said spin-bearing species.

5. The system of claim 4, wherein said magnet comprises an electromagnet.

6. The system of claim 4, wherein said magnet comprises a permanent magnet.

7. The system of claim 4, wherein the combination of said DC magnetic field and said alternating magnetic field provides spin locking of at least a portion of at least one of said spin-bearing species.

8. The system of claim 1, wherein said electrolyte comprises a nonaqueous solvent.

9. The system of claim 8, wherein said electrolyte comprises a room temperature ionic liquid.

10. The system of claim 8, wherein said electrolyte comprises an aprotic solvent.

11. The system of claim 1, wherein said electrolyte comprises uranium.

12. The system of claim 1, wherein said first electrode comprises a liquid metal.

13. A system for isotope selective electrolysis, said system comprising:

an electrolyte comprising a first magnetic ionic species and also comprising a second non-magnetic ionic species that is an isotopologue of said first magnetic ionic species;

at least one first electrode in contact with said electrolyte;

at least one second electrode in contact with said electrolyte;

An alternating current source isolated from said second electrode and coupled to said first electrode for providing an alternating current through said first electrode, wherein said current produces an alternating magnetic field in a portion of electrolyte adjacent to said first electrode, thereby modifying the spin of at least one of the ionic species in said portion of electrolyte adjacent to said first electrode; and,

a current source coupled to said first electrode and to said second electrode for electrolyzing at least a portion of at least one of the two ionic species.

14. The system of claim 1, wherein said alternating magnetic field comprises at least one frequency between 100 kHz and 100 GHz.

15. The system of claim 1, wherein said alternating magnetic field comprises more than one frequency.

16. The system of claim 1, further comprising a magnet for providing a DC magnetic field, wherein said magnetic field manipulates the Zeeman interaction of said spin-bearing species.

17. The system of claim 16, wherein said magnet comprises an electromagnet.

18. The system of claim 16, wherein said magnet comprises a permanent magnet.

19. The system of claim 16, wherein the combination of said DC magnetic field and said alternating magnetic field provides spin locking of at least a portion of at least one of said spin-bearing species.

20. The system of claim 1, wherein said first magnetic ionic species comprises uranium-235.

* * * * *