



US007964301B2

(12) **United States Patent**  
**Fischel et al.**

(10) **Patent No.:** **US 7,964,301 B2**  
(45) **Date of Patent:** **Jun. 21, 2011**

(54) **DYNAMIC ACCELERATED REACTION BATTERIES UTILIZING TAYLOR VORTEX FLOWS**

(75) Inventors: **Halbert Fischel**, Santa Barbara, CA (US); **Philip Michael Lubin**, Santa Barbara, CA (US); **Daniel Timothy Lubin**, Santa Barbara, CA (US)

(73) Assignee: **Global Energy Science, LLC (California)**, Santa Barbara, CA (US)

(\* ) Notice: Subject to any disclaimer, the term of this patent is extended or adjusted under 35 U.S.C. 154(b) by 0 days.

(21) Appl. No.: **12/800,709**

(22) Filed: **May 20, 2010**

(65) **Prior Publication Data**

US 2010/0330460 A1 Dec. 30, 2010

**Related U.S. Application Data**

(60) Provisional application No. 61/220,583, filed on Jun. 26, 2009.

(51) **Int. Cl.**  
**H01M 2/38** (2006.01)

(52) **U.S. Cl.** ..... **429/69; 429/67; 429/68**

(58) **Field of Classification Search** ..... **429/67, 429/68, 69**

See application file for complete search history.

(56) **References Cited**

**U.S. PATENT DOCUMENTS**

5,139,894	A *	8/1992	Mizuno et al.	429/9
5,250,370	A *	10/1993	Faris	429/68
5,332,630	A *	7/1994	Hsu	429/432
5,824,278	A *	10/1998	Yao	423/263
5,830,593	A	11/1998	Nielson	

6,319,293	B1	11/2001	Debe et al.	
6,471,392	B1	10/2002	Holl	
7,488,547	B1	2/2009	Iacovelli	
2002/0155330	A1 *	10/2002	Tanaka	429/19
2002/0168556	A1 *	11/2002	Leboe et al.	429/13
2005/0031517	A1 *	2/2005	Chan	423/263
2006/0062702	A1	3/2006	Hagemeister et al.	
2007/0020142	A1	1/2007	Federspiel et al.	
2008/0149050	A1	6/2008	Shih et al.	

**OTHER PUBLICATIONS**

G.I. Taylor. Stability of a Viscous Liquid Contained between Two Rotating Cylinders, Philosophical Transactions of the Royal Society of London. Series A, Containing Papers of a Mathematical or Physical Character, vol. 223 (1923), pp. 289-343.\*

D. V. Pugh, A. Dursun, and S.G. Corcoran. Formation of nanoporous platinum by selective dissolution of Cu from Cu<sub>0.75</sub>Pt<sub>0.25</sub>, J. Mater. Res. 2003, 18(1), 216-221.\*

Gabe et al, The rotating cylinder electrode: a recent development, Reviews of Applied Electrochemistry 6, J. of Applied Electrochemistry 13 (1983) pp. 3-22.

Gabe et al, The rotating cylinder electrode: its continued development and application, Reviews of Applied Electrochemistry 49, J. of Applied Electrochemistry 28 (1998) pp. 759-780.

Bagotsky, Fundamentals of Chemistry, Second Edition, John Wiley & Sons, Inc., Hoboken, NJ, USA, © 2006, §4.4—Convective Transport, pp. 60-67.

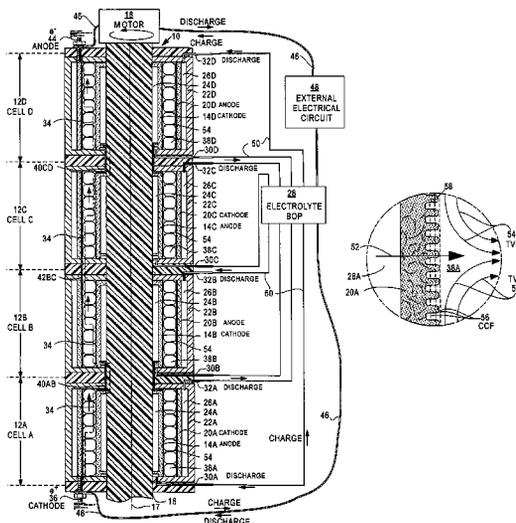
(Continued)

*Primary Examiner* — Jennifer K. Michener  
*Assistant Examiner* — Carlos Barcena  
 (74) *Attorney, Agent, or Firm* — Sheldon L. Epstein; Malcolm J. Chrisholm, Jr.; Pamela J. Curbelo

(57) **ABSTRACT**

Dynamic accelerated reaction batteries (10, 100, 200) with rotating electrodes (14x, 20x, 104x, 106x) or rotating membranes (208) or fuel cells (412C, 412D) that generate Taylor Vortex Flows (54, 122, 228, 454) and Circular Couette Flows (56, 124, 230) in electrolyte chambers (38x, 126x, 206, 438x) are disclosed.

**45 Claims, 9 Drawing Sheets**



OTHER PUBLICATIONS

Newman, *Electrochemical Systems*, Third Edition, John Wiley & Sons, Inc., Hoboken, NJ, USA, © 2004, Chapter 17—Convective-Transport Problems, §17.3-§17.8, pp. 382-399.

Newman, *Electrochemical Systems*, Third Edition, John Wiley & Sons, Inc., Hoboken, NJ, USA, © 2004, Chapter 1—Introduction, §1.4 Transport, pp. 8-18.

Bagotsky, *Fundamentals of Chemistry*, Second Edition, John Wiley & Sons, Inc., Hoboken, NJ, USA, © 2006, Preface to First Edition,

pp. xix-xxi & §1.6 Classification of Electrodes and Electrode Reactions, pp. 12-15.

Newman, *Electrochemical Systems*, Third Edition, John Wiley & Sons, Inc., Hoboken, NJ, USA, © 2004, Preface to the First Edition, pp. xix-xx & §1.2 Thermodynamics and Potential, pp. 4-7.

Pletcher & Walsh, *Industrial Electrochemistry*, Chapman & Hall, London, UK, © 1982, Preface pp. viii-x; Chapter 7, §(d) pp. 346-350 & Chapter 11, p. 543.

\* cited by examiner

FIG. 1A

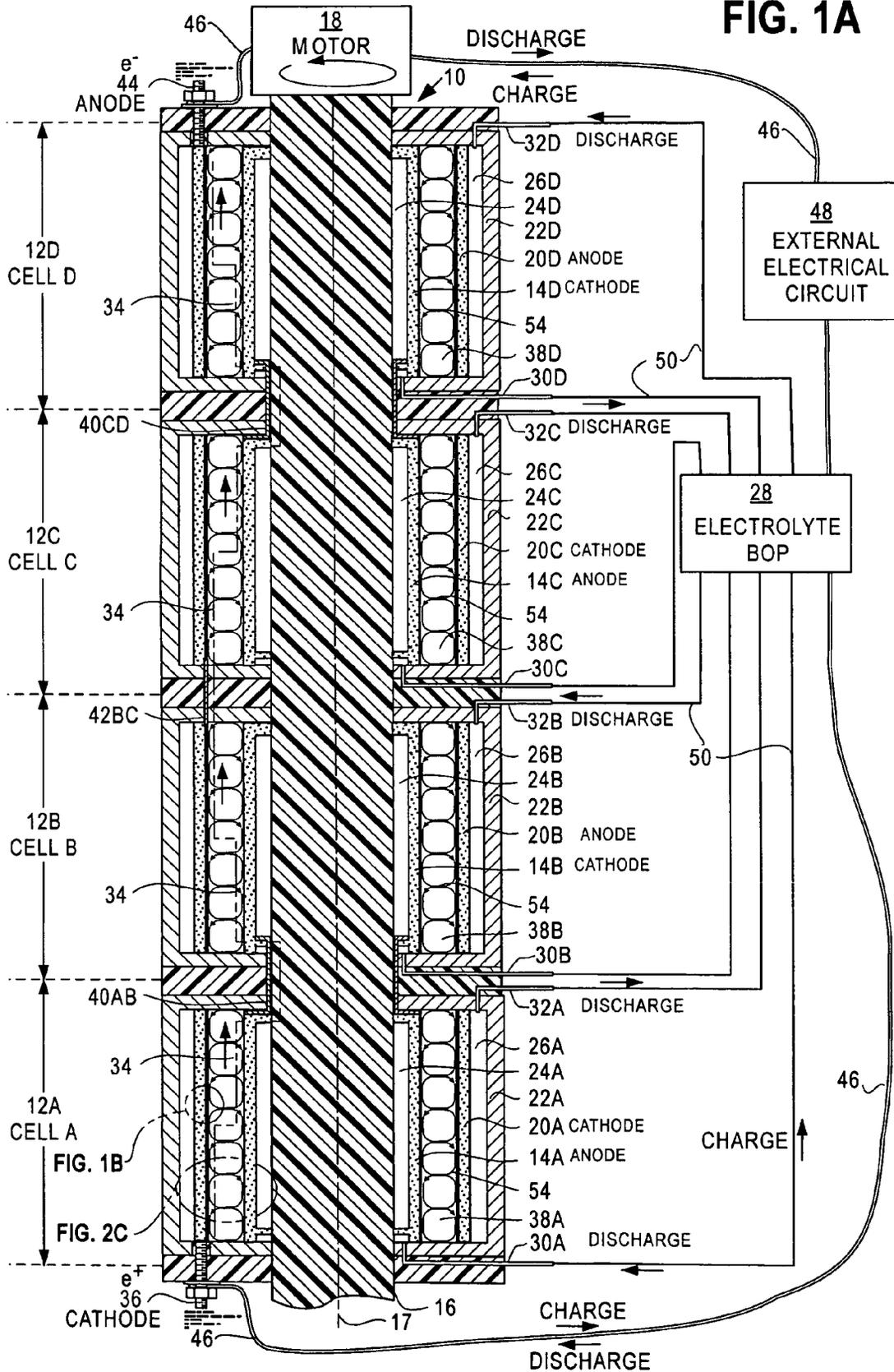


FIG. 2A

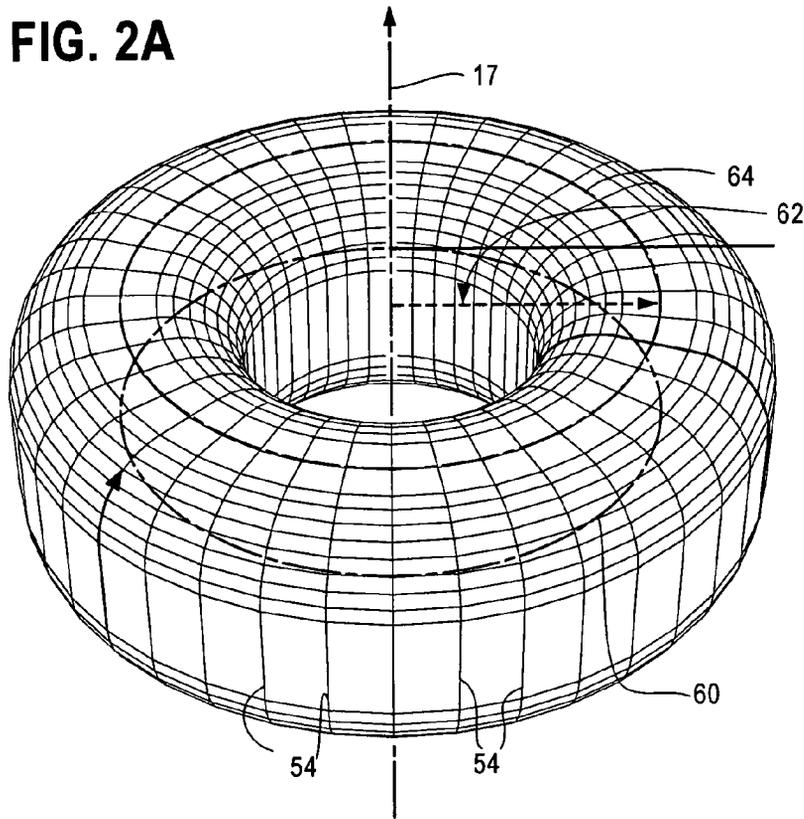


FIG. 2B

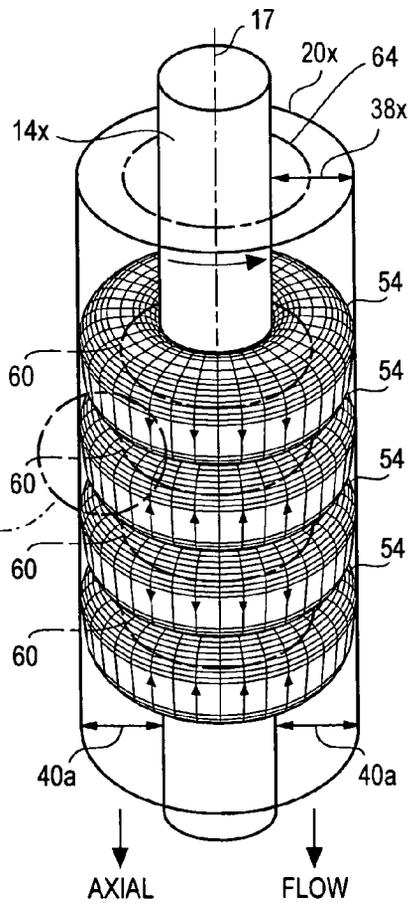


FIG. 1B

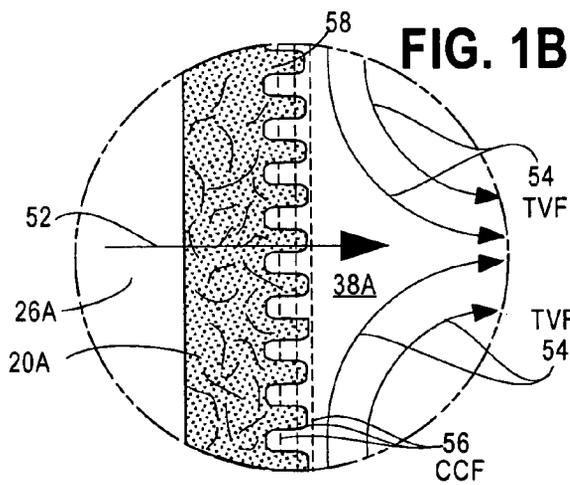


FIG. 2C

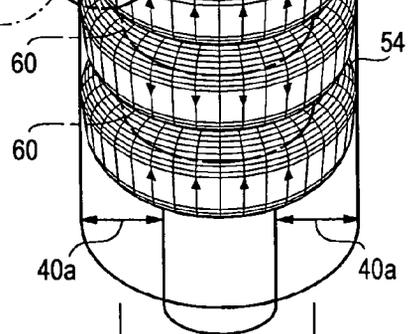


FIG. 2C

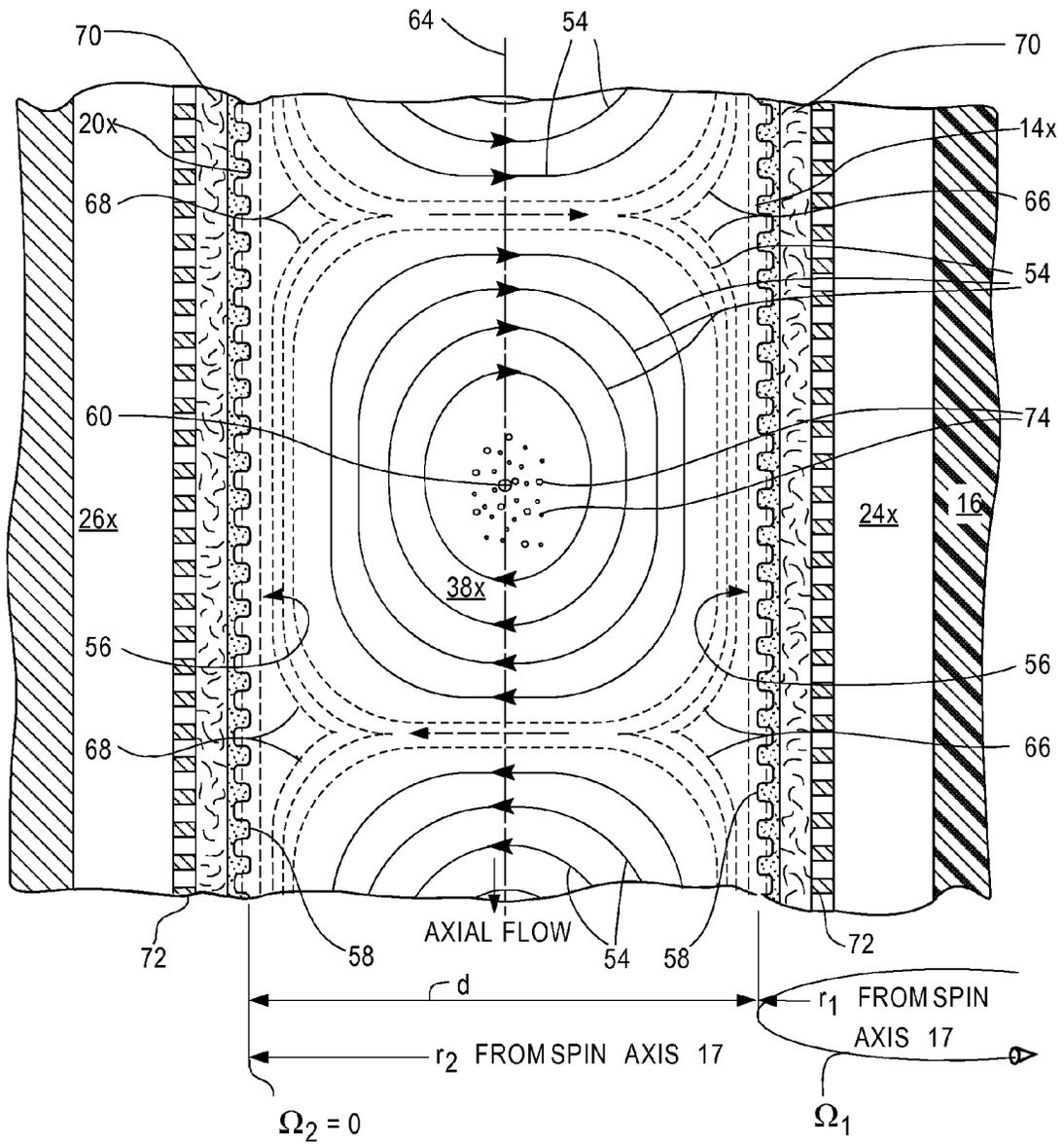


Fig. 2D

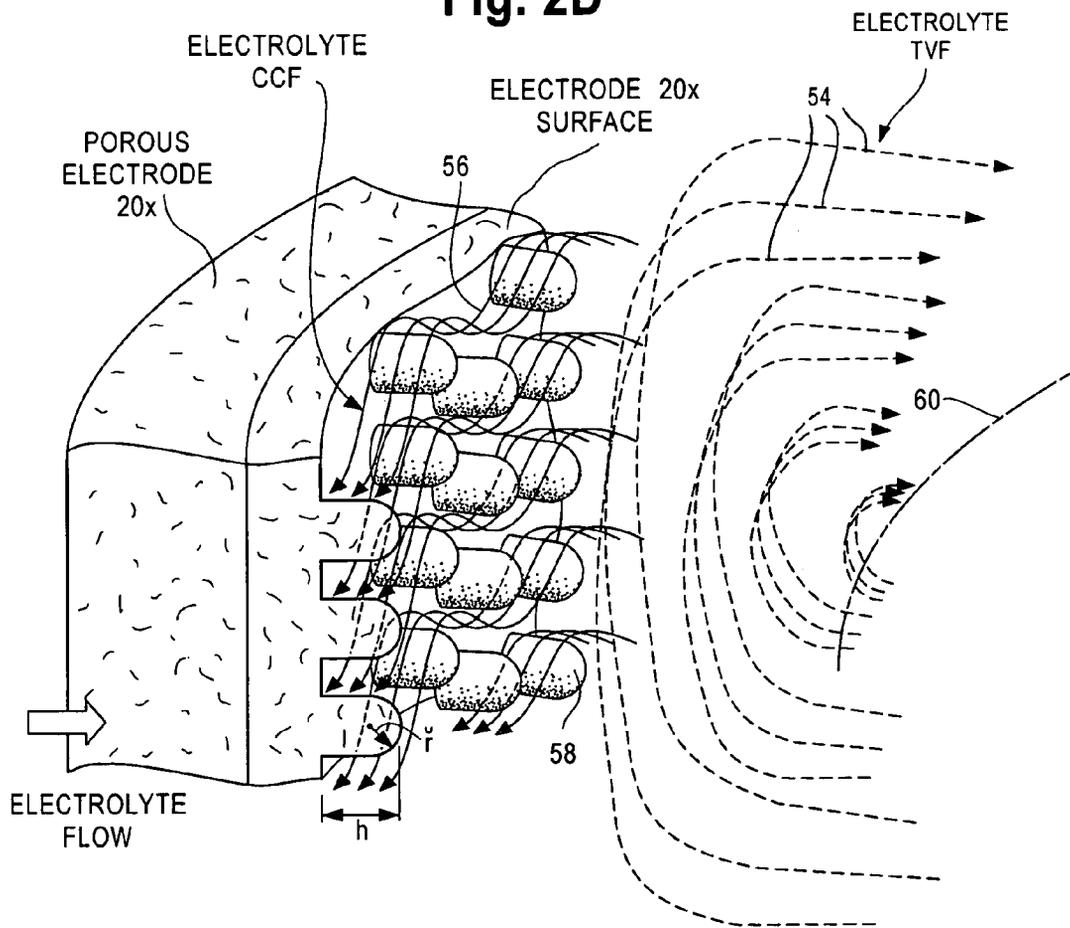
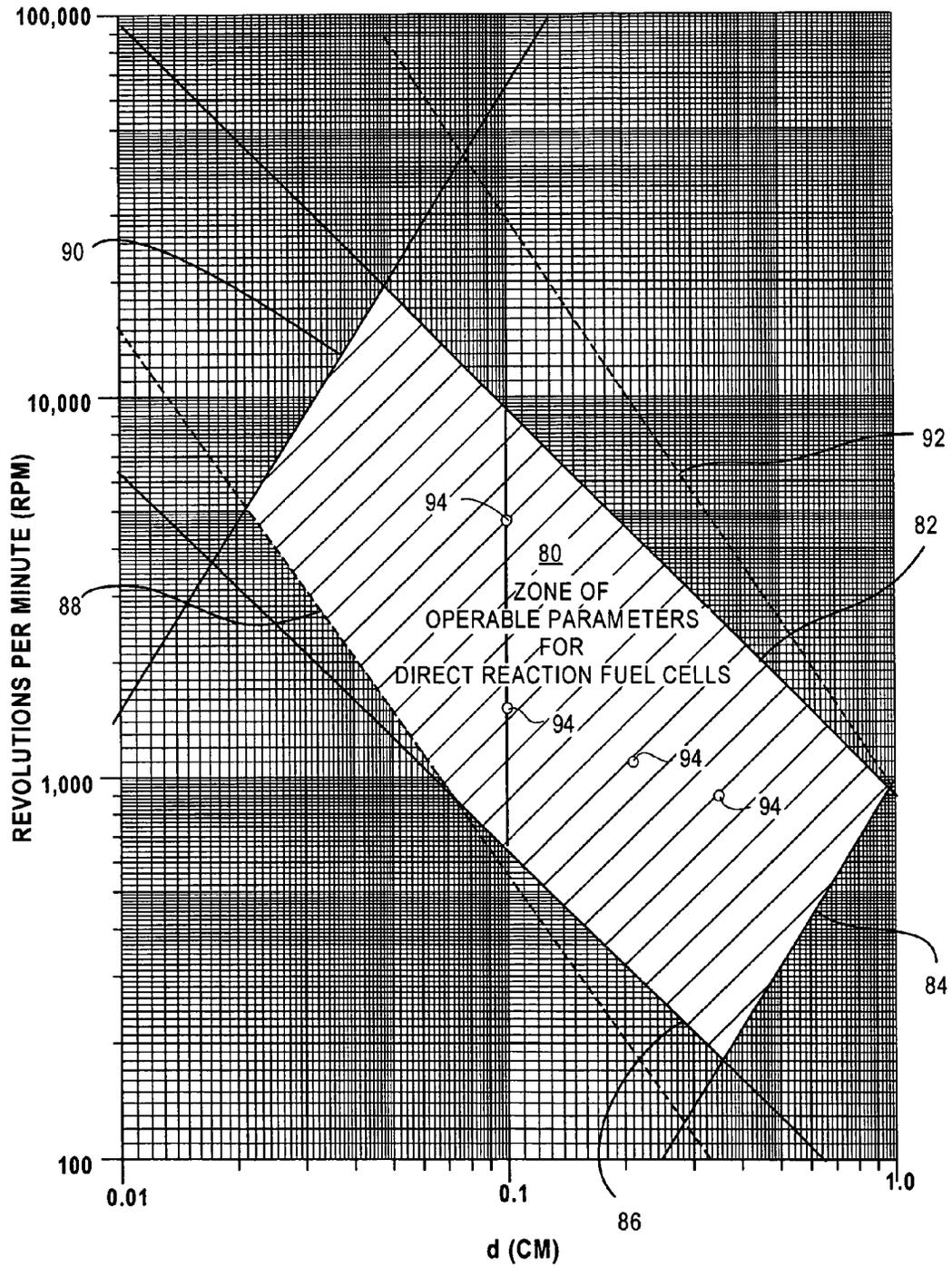


FIG. 3



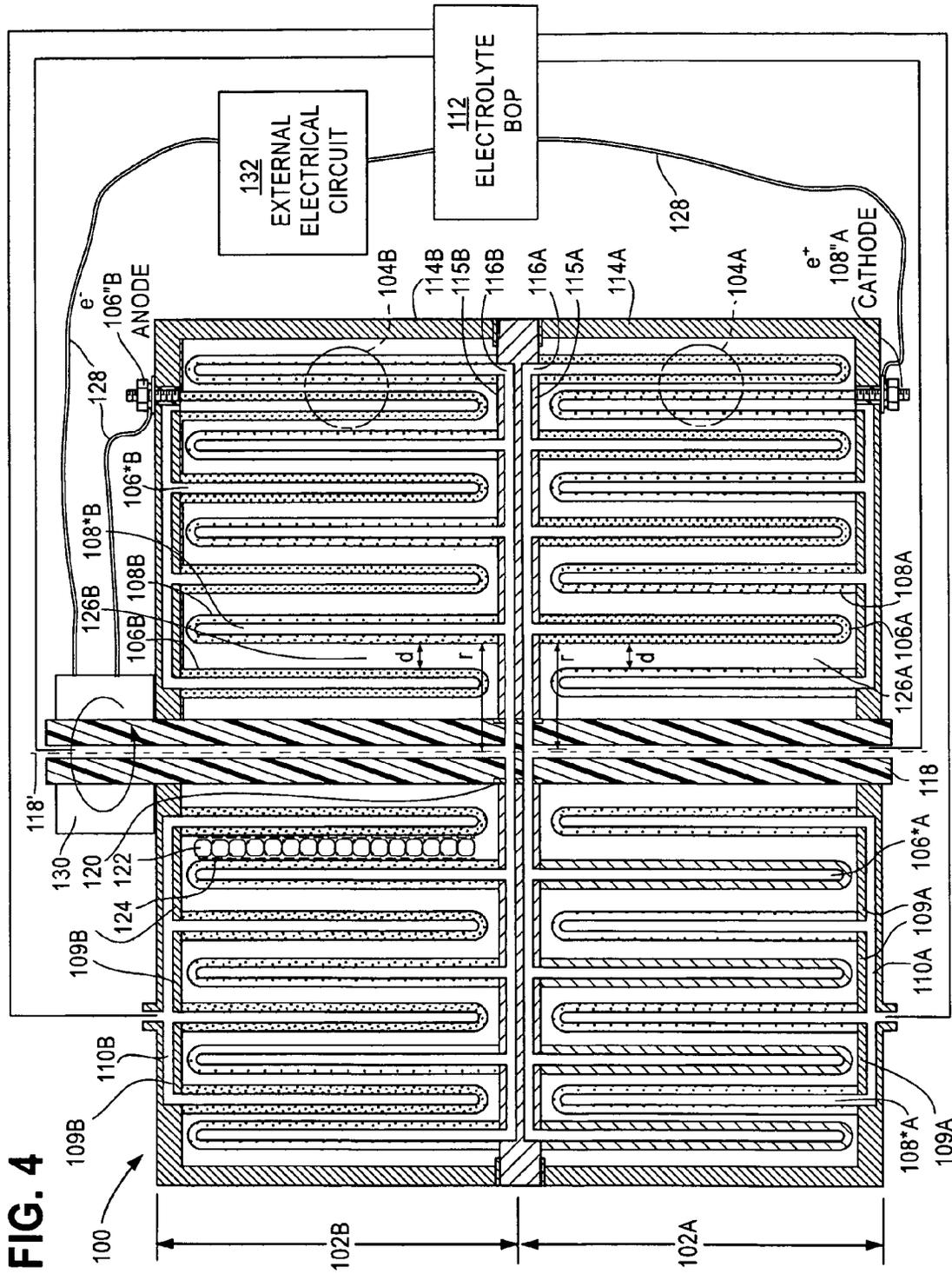


FIG. 5

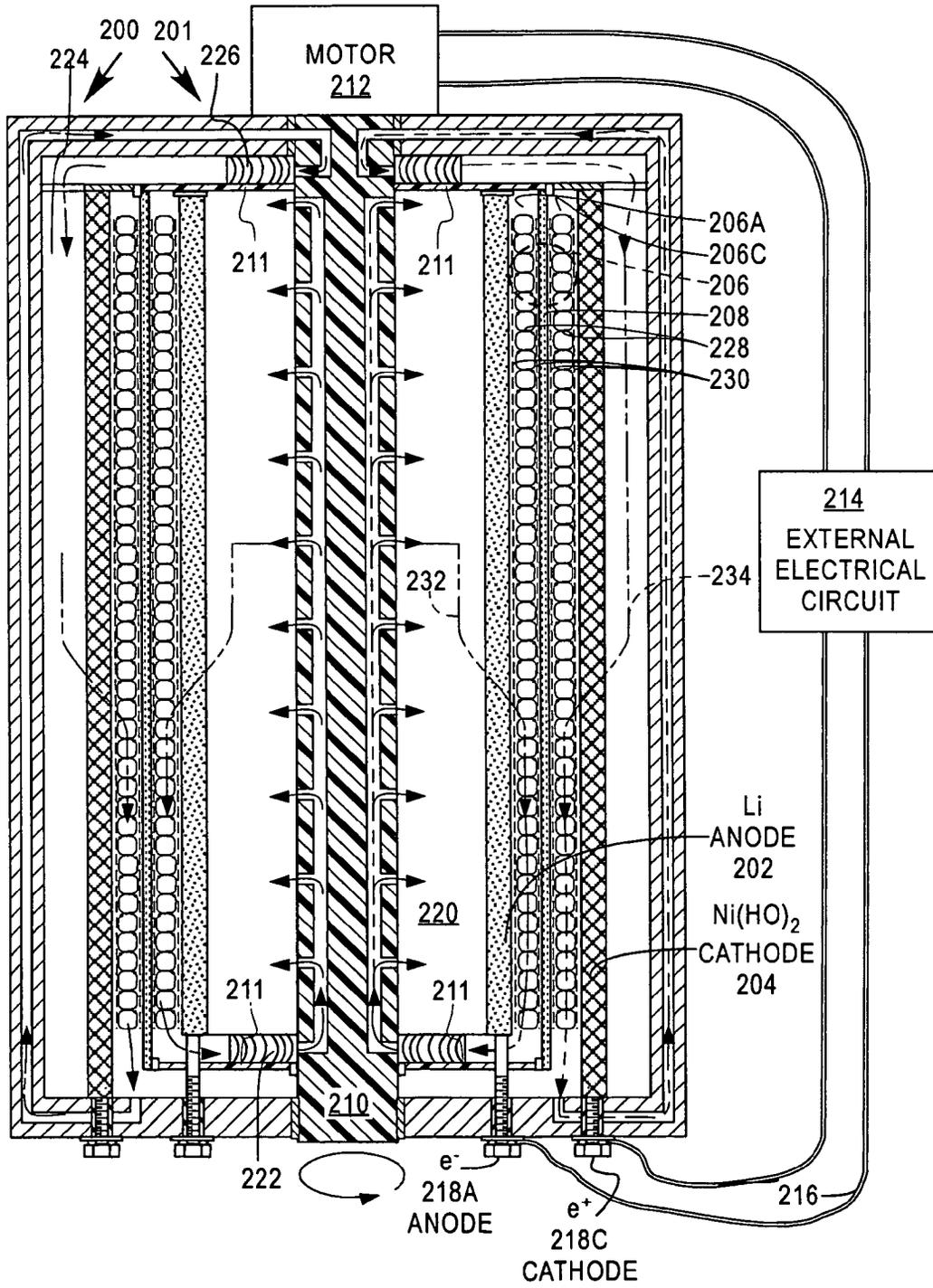


Fig. 6A

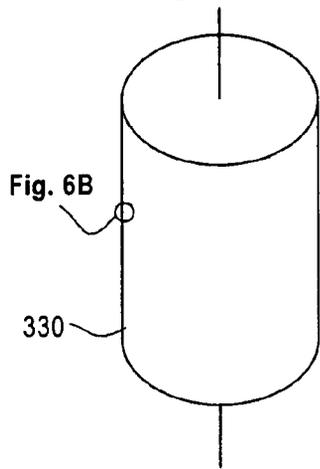


Fig. 6B

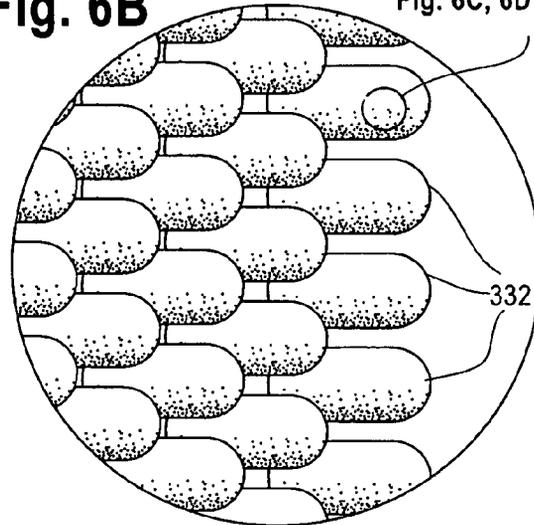


Fig. 6C

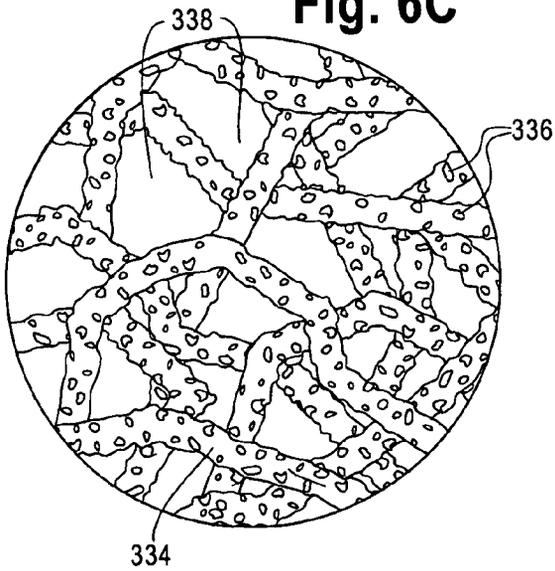


Fig. 6D

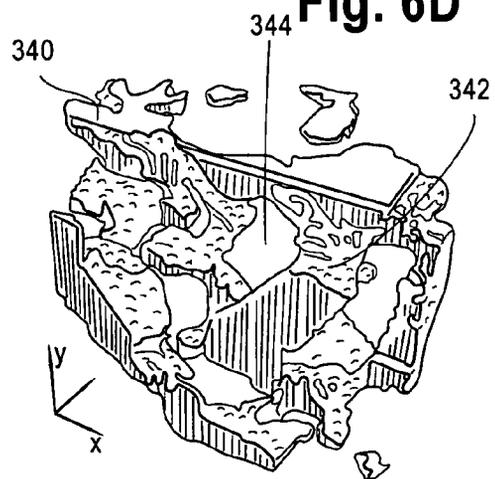
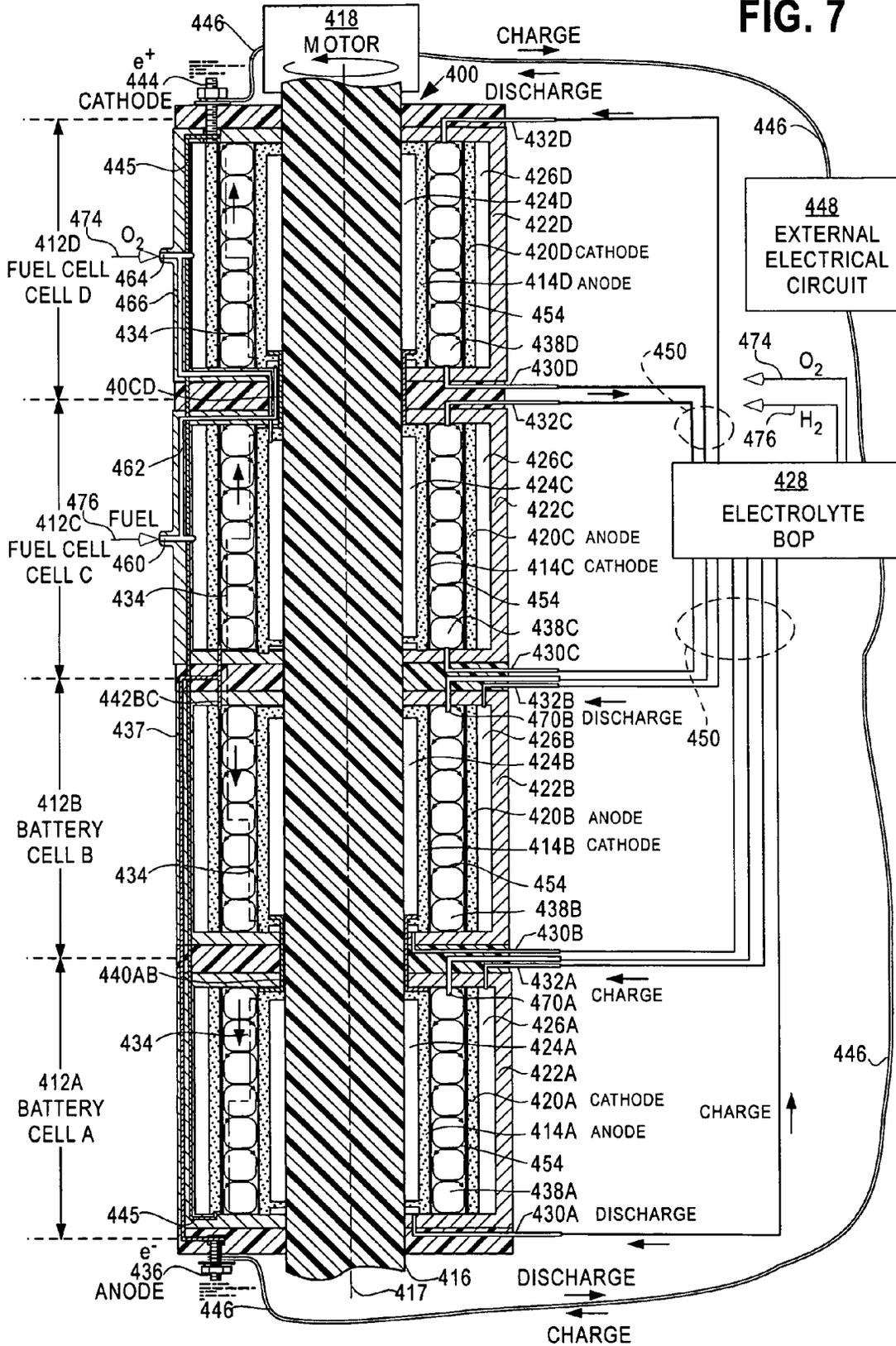


FIG. 7



1

**DYNAMIC ACCELERATED REACTION  
BATTERIES UTILIZING TAYLOR VORTEX  
FLOWS**

CROSS-REFERENCE TO RELATED  
APPLICATIONS

This application claims the benefit of U.S. Provisional Application No. 61/220,583 filed 26 Jun. 2009 by Halbert P. Fischel, which is incorporated herein by reference.

This application, identified as Case E, is related to the following applications of Halbert P. Fischel:

Case A: Electrochemical Cells Utilizing Taylor Vortex Flows, application Ser. No. 12/800,658, U.S. Publication Ser. No. 2010/0330439;

Case B: Fuel Reformers Utilizing Taylor Vortex Flows, application Ser. No. 12/800,710, U.S. Publication Ser. No. 2010/0330445;

Case C: Chemical Process Accelerator Systems Comprising Taylor Vortex Flows, application Ser. No. 12/800,657, U.S. Publication Ser. No. 2010/0329947; and

Case D: Direct Reaction Fuel Cells Utilizing Taylor Vortex Flows, application Ser. No. 12/800,672, U.S. Publication Ser. No. 2010/0330459.

Case A, Case B, Case C, Case D and Case E (this case) were all filed on the same day. All of these applications have been assigned to the same assignee. The other applications are incorporated herein by reference.

STATEMENT REGARDING FEDERALLY  
SPONSORED RESEARCH OR DEVELOPMENT

Not Applicable

THE NAMES OF PARTIES TO A JOINT  
RESEARCH AGREEMENT

Not Applicable

INCORPORATION-BY-REFERENCE OF  
MATERIAL SUBMITTED ON A COMPACT DISK

Not Applicable

BACKGROUND OF INVENTION

1. Field of the Invention

This invention is in the field of electrochemical cells and batteries used to convert chemical energy into electrical energy and having means for providing relative motion between an electrode and an electrolyte—including means for rotating an electrode (U.S. Class 429/67-69, Int. Class H01M), to achieve accelerated chemical reaction rates promoted by Taylor Vortex Flows (TVF) and Circular Couette Flows (CCF).

2. Description of Related Art

Electrochemical cells and batteries (e.g., stacks, piles) have been studied for two-hundred years with objectives of increasing energy density (e.g. Joules/cm<sup>3</sup>, Joules/kg), power density (e.g. Watts/cm<sup>3</sup>, Watts/kg), charge-discharge rate (Amperes/second), terminal voltage, functional operating temperature range and the like. Most improvements in battery technology have been achieved by incremental advances in material selection and fabrication. While these improvements have been important, most of today's cells and batteries struc-

2

turally resemble the Voltaic Pile of 1800 that comprised a stationary anode separated from a stationary cathode by an electrolyte.

Electrochemists have long understood that battery performance would be improved by causing electrolyte to flow between electrodes. The objectives were either to a) accelerate movement of ions between electrodes, b) prevent formation of metallic dendrites on electrodes that could short-circuit cells, and c) prevent or control flooding of electrodes by electrolytes. Some prior art cells or batteries included moving electrodes (e.g., U.S. Pat. No. 4,491,624 to Sarbacher et al, U.S. Pat. No. 4,521,498 to Tamminen and U.S. Pat. No. 4,684,585 to Tamminen) to induce electrolyte flows. However, there is no evidence that any of these cells or batteries is capable of discharging at a rate in excess of 1 ampere/cm<sup>2</sup> of electrode surface for an extended period.

SUMMARY OF THE INVENTION

Dynamic Accelerated Reaction Batteries (DARB) of this invention comprise one or more electrochemical cells capable of fast and high rates of discharge and, for secondary cells, fast and high rates of charge. These attributes are achieved by incorporating one or more rotating electrodes that induce Taylor Vortex Flows (TVF) and Circular Couette Flows (CCF) in electrolytes between anodes and cathodes. Electrodes optimized for use with TVF and CCF and having high current densities and consequent fast charge/discharge rates are described below.

Taylor Vortex Flows (“TVF”—also known as Taylor-Couette Flows) achieve enhanced reaction rates by a) accelerating electrode surface reaction mass-transport, b) capturing reaction products that can degrade electrode surfaces and electrolytes, c) eliminating those degrading reaction products from the cells, d) increasing operating temperature range to reduce overpotentials at electrodes and raise reaction rates and e) concentrate reacting electrode surface area exposure to electrolyte. Chemical and electrochemical cells—including fuel cells and fuel reformers—utilizing these achievements are described in Case A, Case B and Case D.

TVF can occur when a viscous fluid (e.g., electrolyte) is confined in a chamber or gap between two cylinders where one cylinder is rotating with respect to the other. For low differential angular velocities, in terms of circumferential Reynolds number,  $R_{\omega}$ , the flows are steady, purely azimuthal and known as Circular Couette Flows (CCF).

Battery electrodes described here use TVF that automatically generate accelerated, high-shear-rate, stable laminar CCF at the electrode surfaces. These laminar flows increase electrode surface electrical current density by accelerating mass transport of ions and providing means for removing degrading reaction products. Reactive electrodes used in batteries that present high surface area/projected area ratios to electrolytes are described in Case C and below.

In 1923, Sir Geoffrey Ingram Taylor published a groundbreaking paper on stability of the Couette flow. He reported that when the differential velocity, between the opposing cylinder surfaces forming a gap, is increased to a range within observed minimum and maximum speeds, Couette flow becomes unstable. Then, a secondary steady-state is created that is characterized by contra-rotating, axisymmetric, toroidal vortices with unique properties. This secondary steady-state is known as TVF.

Mechanical systems generating TVF are well known in the prior art of particulate filtration. For example, the following U.S. patents, hereby incorporated herein by reference,

describe systems employing TVF for filtering blood without clogging a plasmapheresis membrane filter:

#	Date	Title	Inventor
4,755,300	July 1988	Couette Membrane Filtration Apparatus . . .	Fischel, R et al.
4,808,307	February 1989	Couette Membrane Filtration Apparatus . . .	Fischel, R et al
4,919,817	April 1990	Blood Cell Washing Systems & Method	Schoendorfer et al
5,034,135	July 1991	Blood Fractionation System & Method	Fischel, H.
5,053,121	October 1991	Blood Cell Washing System & Methods	Schoendorfer et al
5,194,145	March 1993	Method . . . For Separation of Matter . . .	Schoendorfer
5,376,263	December 1994	Pump Control Apparatus . . . Rotating . . .	Fischel, H.
5,464,534	November 1995	Blood Fractionation System & Method	Fischel, H.
5,738,792	April 1998	Method For Separation of Matter . . .	Schoendorfer
5,783,085	July 1998	Blood Fractionation Method	Fischel, H.

Particulate filters are readily distinguished from batteries and cells because 1) the filters lack electrodes and 2) the direction of flow of reactant fluids is diametrically opposite to that of batteries and cells.

In particulate filters such as these blood filters, a fluid, such as blood, containing a suspended particulate, such as blood cells, is pumped through a gap between opposing cylinder walls. One wall, usually the outer, is solid while the other is porous. The porous wall usually incorporates filter media and rotates within the outer wall. Fluid penetrates the filter media on the inner wall where TVF-accelerated high-shear-rate laminar flow prevents particulates from entering and clogging the filter media pores. TVF trap the particulates and transport them to an exit from the gap to be purged from the system.

Batteries and cells of this invention comprise different structures and employ TVF in a different manner. For example, cells incorporate porous anode and cathode electrodes separated by electrolyte. In embodiments described below, the electrolyte penetrates each of the electrodes in reversible directions for charge/discharge cycles, respectively, by moving in cross flow through a gap between the electrodes.

This disclosure describes systems and methods for management of any type of fluid electrolyte in electrochemical cells and batteries. Cells and batteries of this invention have ionic conductivities that are orders of magnitude greater than prior art cells and batteries so that ionic transfer time between electrodes that have been a mass transport limitation of electrical-current-producing chemical reactions now are negligible.

For cells and batteries incorporating membranes (e.g., lithium cells using LISICON™ membranes to separate an organic anolyte from an aqueous catholyte, such as U.S. Pat. No. 5,525,442 to Shuster), the dominant limitation on ionic conductivity is the rate at which ions can migrate through the membrane. This rate can be increased by creating high-shear-rate laminar flow at the surfaces of the membrane. Laminar CCF are in direct contrast with the prior art that teaches that turbulent flow is beneficial in reducing dendrite formation and increasing electrode activity.

The prior art also teaches that in order to maintain laminar flow, fluid (e.g., electrolyte) velocities must be kept sufficiently low so that the critical turbulent Reynolds Number,  $Re$

(not  $R_c$ ), is not exceeded. The present invention of this disclosure succeeds because its cells have electrolyte velocities that result in much higher circumferential Reynolds Numbers,  $R_c$ , and high laminar shear rates stabilized by generated TVF. Because a requirement for laminar flow can be achieved at much higher laminar shear rates using TVF, as taught here, cells and batteries of this invention achieve multiple orders of magnitude higher current densities than non-TVF laminar flow cells and batteries.

The controlling factor for laminar shear rates is the minimum value of the hydro-dynamically defined Taylor number,  $T_c$  above which desirable energetic vortices are fully established within the electrolyte. Axial Poiseuille-type flow can be further imposed by virtue of recirculation. Also, there is a strong dependence of the critical  $T_c$  on the strength of the latter flow by way of its characteristic axial-flow Reynolds number,  $R_a$ .

Furthermore, there is a requirement to maintain laminar flow at electrode surfaces while promoting TVF within the electrolyte. Care must be taken to restrict the circumferential Reynolds number,  $R_c$ , to non-turbulent values. A set of distinct variables define a particular range of permissible operating parameters. These variables include predetermined ranges of: 1) temperature, 2) kinematic viscosity of the fluids being employed, 3) respective rates of recirculation, 4) angular rotation speed, 5) surface characteristics of the electrodes and 6) physical dimensions of the cell.

Given appropriate operating parameters, neither gaseous reaction products nor particle precipitates of the redox reaction at the electrodes can penetrate the TVF. Only redox-product ions in solution can enter the electrolyte and cross rapidly to the opposite electrode, which is an important feature of electrochemical cells utilizing TVF in electrolyte.

The present invention is a robust electrochemical cell or battery for providing electrical energy containing a fluid electrolyte between relatively rotating, co-axial cylindrical electrodes that cause Taylor Vortex Flows (TVF) and Circular Couette Flows (CCF) within the electrolyte. Where a membrane is required for separation of incompatible electrolytes, TVF and CCF enhance ion transport through the membrane.

It is therefore a first object of the present invention to provide a robust battery utilizing TVF and CCF to provide greatly increased mass transport of ions within the electrolyte and between electrodes.

A second object of the present invention is to provide a robust battery utilizing TVF and CCF to utilize greatly increased active or exposed electrode areas for a given cell volume exposed to electrolytes.

A third object of the present invention is to provide a battery that does not require any membrane disposed between electrodes.

A fourth object of the present invention is to provide a battery that can utilize a membrane, as when required for separating incompatible electrolytes, without increasing internal cell resistance.

A fifth object of this invention is to provide a TVF-CCF battery that is mechanically and electrically compatible with fuel cells, such as TVF-CCF fuel cells of the types disclosed in Case A and Case D.

A sixth object of this invention is to provide DARB that create dynamic flows within electrolytes to induce exceptionally high rates of mass transport to and from several redox chemical reactions.

A seventh object of this invention is to provide DARB with low mass-transport-limiting and surface-limiting electrode

5

characteristics and dynamic forced convective flow of electrolyte through electrodes that have very high surface area volumetric concentrations.

An eighth object of this invention is to provide DARB in synergistic combination with a fuel cell.

These and other objects of this invention are more fully set forth in the following description of a preferred embodiment of this invention.

#### BRIEF DESCRIPTIONS OF THE SEVERAL VIEWS OF THE DRAWINGS

FIG. 1A is a fragmentary cross-sectional view of a first preferred embodiment of Dynamic Accelerated Reaction Batteries comprising a series of connected cells.

FIG. 1B is a magnified view of a portion of FIG. 1A showing a portion of one of the battery's electrodes.

FIG. 2A, FIG. 2B, FIG. 2C and FIG. 2D are detailed illustrations of a Taylor Vortex Flows and Circular Couette Flows in Dynamic Accelerated Reaction Battery.

FIG. 3 is a log-log graph of Revolutions Per Minute (RPM) as a function of gap width,  $d$ , for a range or zone of operable conditions.

FIG. 4 is a fragmentary cross-sectional view of a second preferred embodiment of a Dynamic Accelerated Reaction Battery comprising interdigitated electrodes.

FIG. 5 is a fragmentary cross-sectional view of a third preferred embodiment of a Dynamic Accelerated Reaction Battery for comprising an ion-permeable membrane for use with incompatible electrolytes.

FIG. 6A is a perspective view of an electrode of this invention.

FIG. 6B is a perspective and magnified view of the surface of the electrode of FIG. 6A.

FIG. 6C is a highly-magnified view of one embodiment of the electrode surface of FIG. 6B.

FIG. 6D is a computer-simulated view of another electrode surface.

FIG. 7 is a fragmentary cross-sectional view of a Dynamic Accelerated

Reaction Battery incorporating a Direct Reaction Fuel Cell.

#### DETAILED DESCRIPTION OF THE INVENTION

##### Dynamic Accelerated Reaction Battery

##### First Embodiment

##### Four Cell Stack

FIG. 1A is a fragmentary cross-section view of a Dynamic Accelerated Reaction Battery 10. FIG. 1A resembles FIG. 6 of Case D, which is a cross-section view of a TVF/CCF fuel cell. While the battery 10 and the fuel cell of Case D both convert their potential energies to electrical energy and share many similarities relating to TVF and CCF, they operate differently. DARB and most conventional batteries store potential energy in their electrodes or dual electrolytes while fuel cells store potential energy in their fuels. As a result, batteries and fuel cells have different energy densities, power densities, terminal voltages and currents.

The battery 10 comprises four cells CELL A-12A, CELL B-12B, CELL C-12C and CELL D-12D. Each cell contains a rotating electrode  $14_x$  where  $x$  stands for one of the letters A, B, C or D that is associated with one of the four cells  $12_x$ . The electrodes  $14_x$  are fixed to a shaft 16 that rotates about spin

6

axis 17 and is driven by an electric motor 18. Each cell also contains a fixed electrode  $20_x$  that is secured to frame  $22_x$ . The motor 18 rotates so that so that there is relative motion between one electrode  $14_x$ ,  $20_x$  of one polarity and another electrode  $20_x$ ,  $14_x$  of another polarity. In this embodiment, the electrode 14A rotates within the electrode 20A, the electrode 14B rotates with the electrode 20B, the electrode 14C rotates with the electrode 20C and the electrode 14D rotates within the electrode 20D. Each frame  $22_x$  is mechanically coupled to and electrically insulated from adjacent frame(s)  $22_x$ .

Cylindrical electrodes  $14_x$  and  $20_x$  are shown in the drawings as right-circular and coaxial; however, these attributes are not a requirement and other cylinder-like geometries (e.g., elliptical, conical, hyperbolic, irregular, different axes) may be employed. Also, it is practical to build DARB 10 where the inner electrodes  $14_x$  are fixed and the outer electrodes  $20_x$  rotate or both electrodes  $14_x$ ,  $20_x$  rotate at different speeds or directions. All that is required is that one of the cylinder-like electrodes  $14_x$  and  $20_x$  rotates relative to the other so that there is a relative, rotational velocity difference greater than zero between the cylindrical electrodes  $14_x$  and  $20_x$ .

Rotating electrodes 14A and 14C are anodes. Rotating electrodes 14B and 14D are cathodes. In this embodiment all of the electrodes  $14_x$  are fixed to and rotated by the shaft 16, which is common to all for rotating the electrodes  $14_x$ ; however a common shaft is not a requirement. Fixed electrodes 20A and 20C are cathodes. Fixed electrodes 20B and 20D are anodes. In this embodiment, all of the electrodes  $20_x$  are fixed to their frames  $22_x$ , which are mechanically coupled together; however, that is not a requirement. As will be described, the cells  $12_x$  are connected in an electrical series circuit; but, parallel and combination circuits are also practical.

Each of the cells  $12_x$  has a rotating electrode manifold  $24_x$  and a fixed electrode manifold  $26_x$ . Each of the manifolds  $24_x$  and  $26_x$  is connected to electrolyte Balance of Plant (BOP) 28 through first and second electrolyte ducts  $30_x$  and  $32_x$  so that during discharge electrolyte can flow through anode electrodes 14A, 20B, 14C and 20D through electrolyte chambers 38X and back to BOP 28 through electrodes 20A, 14B, 20C and 14D. The electrodes  $14_x$  are rotated by the motor 18 so that there is relative motion between one electrode of one polarity and another electrode of another polarity as well as relative motion of the electrodes  $14_x$  with respect to the electrolyte.

An electric circuit—illustrated in part as dashed line 34—is then formed that starts at battery cathode  $e^+$  terminal 36 that is electrically connected to fixed electrode 20A. The circuit follows near circuit dashed line 34 up fixed electrode 20A, across electrolyte chamber 38A, up rotating electrode 14A to copper alloy sleeve busbar 40AB that provides a low-electrical-impedance connection between anode rotating electrode 14A and cathode rotating electrode 20B. The busbar 40AB is secured to and electrically insulated from the rotating shaft 16. The busbar 40AB is also insulated (not shown) from electrolyte in the chambers 38A and 38B to protect it from chemical attack.

The electric circuit continues up rotating electrode 14B, across electrolyte chamber 38B and up fixed anode electrode 20B to copper alloy busbar 42BC, which is secured to and insulated from frames 22B and 22C. The busbar 42BC provides a low-electrical-impedance connection between anode fixed electrode 20B and cathode fixed electrode 20C. The electric circuit continues up electrode 20C, across electrolyte chamber 38C to rotating anode electrode 14C and to busbar sleeve 40CD, which is essentially identical to busbar sleeve 40AB. The busbar sleeve 42CD provides a low-electrical-





























