



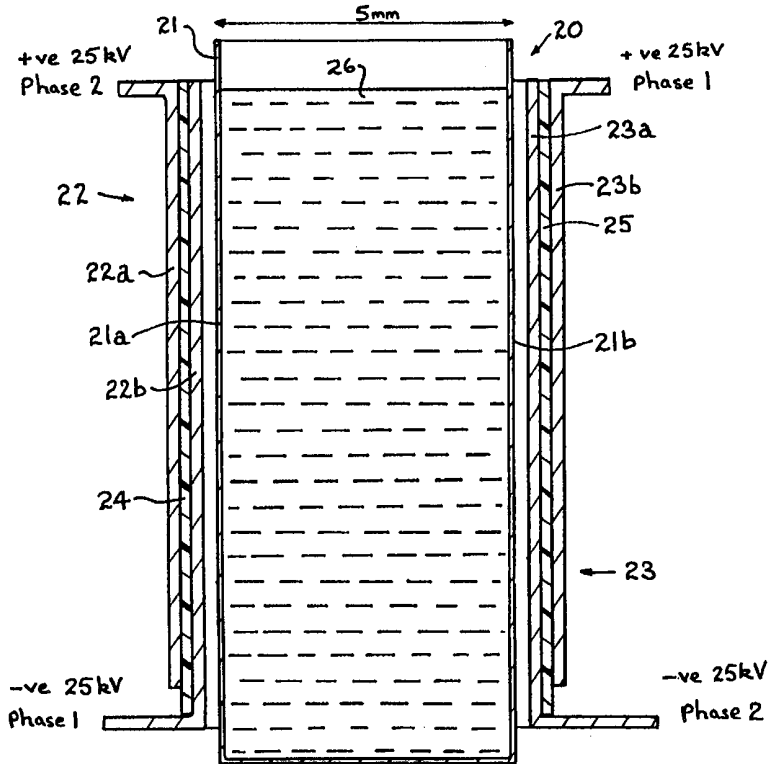
INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<p>(51) International Patent Classification ⁶ : B01J 19/08, C01B 3/04, B01J 7/02</p>	<p>A1</p>	<p>(11) International Publication Number: WO 98/46349 (43) International Publication Date: 22 October 1998 (22.10.98)</p>
<p>(21) International Application Number: PCT/GB98/01099 (22) International Filing Date: 15 April 1998 (15.04.98) (30) Priority Data: 9707760.6 17 April 1997 (17.04.97) GB (71) Applicants (for all designated States except US): DAVIES, Christopher, John [GB/GB]; Westgate House, Dedham, Colchester, Essex CO7 6HJ (GB). DAVIES, Caroline, Jane [GB/GB]; Westgate House, Dedham, Colchester, Essex CO7 6HJ (GB). (71)(72) Applicant and Inventor: ECCLES, Christopher, Robert [GB/GB]; Westgate House, Dedham, Colchester, Essex CO7 6HJ (GB). (74) Agent: NEWBY, Martin, John; J.Y. & G.W. Johnson, Kingsbourne House, 229-231 High Holborn, London WC1V 7DP (GB).</p>	<p>(81) Designated States: AL, AM, AT, AT (Utility model), AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, CZ (Utility model), DE, DE (Utility model), DK, DK (Utility model), EE, EE (Utility model), ES, FI, FI (Utility model), GB, GE, GH, GM, GW, HU, ID, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SK (Utility model), SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).</p> <p>Published <i>With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i></p>	

(54) Title: FRACTURE CELL APPARATUS

(57) Abstract

Fracture cell apparatus including a capacitive fracture cell (20) comprising a container (21) having walls (21a, 21b) made of non-electrically conducting material for containing a liquid dielectric (26), and spaced apart electrode means (22, 23) positioned outside the container (21) with the liquid dielectric (26) between the electrode means, and means (1-8a, 8b) for applying positive and negative voltage pulses to each of the electrode means (22, 23). In use, whenever one of a positive voltage pulse and a negative voltage pulse is applied to one of the two electrode means, the other of a positive voltage pulse and a negative voltage pulse is applied to the other of the two electrode means, thereby creating an alternating electric field across the liquid dielectric to cause fracture of the liquid dielectric (26). The invention also relates to a method of generating gas, preferably the decomposition of water into gaseous hydrogen and oxygen.



FOR THE PURPOSES OF INFORMATION ONLY

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

AL	Albania	ES	Spain	LS	Lesotho	SI	Slovenia
AM	Armenia	FI	Finland	LT	Lithuania	SK	Slovakia
AT	Austria	FR	France	LU	Luxembourg	SN	Senegal
AU	Australia	GA	Gabon	LV	Latvia	SZ	Swaziland
AZ	Azerbaijan	GB	United Kingdom	MC	Monaco	TD	Chad
BA	Bosnia and Herzegovina	GE	Georgia	MD	Republic of Moldova	TG	Togo
BB	Barbados	GH	Ghana	MG	Madagascar	TJ	Tajikistan
BE	Belgium	GN	Guinea	MK	The former Yugoslav Republic of Macedonia	TM	Turkmenistan
BF	Burkina Faso	GR	Greece	ML	Mali	TR	Turkey
BG	Bulgaria	HU	Hungary	MN	Mongolia	TT	Trinidad and Tobago
BJ	Benin	IE	Ireland	MR	Mauritania	UA	Ukraine
BR	Brazil	IL	Israel	MW	Malawi	UG	Uganda
BY	Belarus	IS	Iceland	MX	Mexico	US	United States of America
CA	Canada	IT	Italy	NE	Niger	UZ	Uzbekistan
CF	Central African Republic	JP	Japan	NL	Netherlands	VN	Viet Nam
CG	Congo	KE	Kenya	NO	Norway	YU	Yugoslavia
CH	Switzerland	KG	Kyrgyzstan	NZ	New Zealand	ZW	Zimbabwe
CI	Côte d'Ivoire	KP	Democratic People's Republic of Korea	PL	Poland		
CM	Cameroon	KR	Republic of Korea	PT	Portugal		
CN	China	KZ	Kazakistan	RO	Romania		
CU	Cuba	LC	Saint Lucia	RU	Russian Federation		
CZ	Czech Republic	LI	Liechtenstein	SD	Sudan		
DE	Germany	LK	Sri Lanka	SE	Sweden		
DK	Denmark	LR	Liberia	SG	Singapore		
EE	Estonia						

- 1 -

Fracture Cell Apparatus

This invention relates to fracture cell apparatus and to a method of generating fuel gas from such fracture cell apparatus. In particular, but not exclusively, the invention relates to an apparatus and method for providing fuel gas from water.

Conventionally, the principal methods of splitting a molecular species into its component atomic constituents have been either purely chemical or purely electrolytic. Purely chemical reactions always involve "third-party" reagents and do not involve the interaction of (1) an applied external electrical influence, and (2) a simple substance. Conventional electrolysis involves the passage of an electric current through a medium (the electrolyte), such current being the product of ion-transits between the electrodes of the cell. When ions are attracted towards either the cathode or the anode of a conventional electrolytic cell, they either receive or donate electrons on contact with the respective electrode. Such electron exchanges constitute the current during electrolysis. It is not possible to effect conventional electrolysis to any useful degree without the passage of this current; it is an a priori feature of the process.

A number of devices have recently been described which purport to effect "fracture" of, particularly, water by means of resonant electrostatic phenomena. In particular one known device and process for producing oxygen and hydrogen from water is disclosed in US-A-4936961. In this known device a so-called fuel cell water "capacitor" is provided in which two concentrically arranged spaced apart "capacitor" plates are positioned in a container of water, the water contacting, and serving as the dielectric between, the "capacitor" plates. The "capacitor" is in effect a charge-dependent resistor which begins to conduct after a small displacement current begins to flow. The "capacitor" forms part of a resonant charging circuit that includes an

- 2 -

inductance in series with the "capacitor". The "capacitor" is subjected to a pulsating, unipolar electric charging voltage which subjects the water molecules within the "capacitor" to a pulsating electric field between the capacitor plates. The "capacitor" remains charged during the application of the pulsating charging voltage causing the covalent electrical bonding of the hydrogen and oxygen atoms within the water molecules to become destabilised, resulting in hydrogen and oxygen atoms being liberated from the molecules as elemental gases.

Such known fracture devices have, hitherto, always featured, as part of their characteristics, the physical contact of a set of electrodes with the water, or other medium to be fractured. The primary method for limiting current flow through the cell is the provision of a high-impedance power supply network, and the heavy reliance on the time-domain performance of the ions within the water (or other medium), the applied voltage being effectively "switched off" in each cycle before ion-transit can occur to any significant degree.

In use of such a known system, there is obviously an upper limit to the number of ion-migrations, electron-captures, and consequent molecule-to-atom disruptions which can occur during any given momentary application of an external voltage. In order to perform effectively, such devices require sophisticated current-limiting and very precise switching mechanisms.

A common characteristic of all such known fracture devices described above, which causes them to behave as though they were conventional electrolysis cells at some point in time after the application of the external voltage, is that they have electrodes in actual contact with the water or other medium.

- 3 -

The present invention seeks to provide an alternative method of producing fracture of certain simple molecular species, for example water.

According to one aspect of the present invention
5 there is provided fracture cell apparatus including a capacitive fracture cell comprising a container having walls made of non-electrically conducting material for containing a liquid dielectric, and spaced apart electrode means positioned outside the container with the liquid dielectric
10 between the electrode means, and means for applying positive and negative voltage pulses to each of the electrode means so that, whenever one of a positive voltage pulse and a negative voltage pulse is applied to one of the two electrode means, the other of a positive voltage pulse and
15 a negative voltage pulse is applied to the other of the two electrode means, thereby creating an alternating electric field across the liquid dielectric to cause fracture of the liquid dielectric.

In apparatus according to the invention the electrode
20 means do not contact the liquid dielectric which is to be fractured or disrupted. The liquid to be fractured is the simple dielectric of a capacitor. No purely ohmic element of conductance exists within the fracture cell and, in use, no current flows due to an ion-carrier mechanism within the
25 cell. The required fracture or disruption of the liquid dielectric is effected by the applied electric field whilst only a simple displacement current occurs within the cell.

Preferably the liquid dielectric comprises water, e.g. distilled water, tap water or deuterated water.

30 Conveniently each electrode means comprises a bipolar electrode.

Suitably the means for alternately applying positive and negative pulses provides step voltages alternately to the two electrode means with a short period of time during

- 4 -

each charge voltage cycle in which no step voltage is applied to either electrode. Typically step voltages in excess of 15 kV, typically about 25 kV, on either side of a reference potential, e.g. earth, are applied to the electrode means. In effect trains of pulses having alternating positive and negative values are applied to the electrode means, the pulses applied to the different electrode means being "phase shifted". In the case where each electrode means comprises a bipolar electrode, each bipolar electrode comprising first and second electrode "plates" electrically insulated from each other, a train of positive pulses is arranged to be applied to one electrode plate of each bipolar electrode and a train of negative pulses is arranged to be applied to the other electrode plate of each bipolar electrode. One electrode plate of one bipolar electrode forms a first set with one electrode plate of the other bipolar electrode and the other electrode plate of the one bipolar electrode forms a second set with the other electrode plate of the other bipolar electrode. For each set, a positive pulse is applied to one electrode plate and a negative pulse is applied simultaneously to the other electrode plate. By alternately switching the application of positive and negative pulses from one to the other set of electrode plates, an "alternating" electric field is generated across the dielectric material contained in the container. The pulse trains are synchronised so that there is a short time interval between the removal of pulses from one electrode plate set and the application of pulses to the other electrode plate set.

According to another aspect of the present invention there is provided a method of generating gas comprising applying positive and negative voltage pulses alternately to electrode means positioned either side of, but not in contact with, a liquid dielectric, the voltage pulses being applied so that, whenever one of a positive voltage pulse and a negative voltage pulse is applied to one of the two electrode means, the other of a positive voltage pulse and a negative voltage pulse is applied to the other of the two

- 5 -

electrodes means, the applied voltage pulses generating an alternating electric field across the liquid dielectric causing fracture of the liquid dielectric into gaseous media.

5 Preferably voltages of at least 15 kV, e.g. 25 kV, either side of a reference value, e.g. earth, are applied across the liquid dielectric to generate the alternating electric field.

10 An embodiment of the invention will now be described by way of example only, with particular reference to the accompanying drawings, in which:

Figure 1 is a circuit diagram of fracture cell apparatus according to the invention;

15 Figure 2 shows in more detail a part of the circuit diagram of Figure 1;

Figures 3a to 3d show different waveforms at various parts of the circuit diagram of Figure 1;

20 Figure 4 is a schematic diagram of a fracture cell for use in fracture cell apparatus according to the invention, and

Figure 5 shows trains of pulses applied to electrode means of the fracture cell apparatus according to the invention.

25 If a large electric field is applied across a pair of electrode plates positioned either side of a cell containing water, disruption of the water molecules will occur. Such disruption yields hydrogen nuclei and HO^- ions. Such a molecular disruption is of little interest in terms of obtaining a usable result from the cell. A proton-rich zone
30 exists for as long as the field exists and quickly

- 6 -

reestablishes equilibrium ion-product when the field is removed.

One noticeable side-effect, however, is that the hydroxyl ions (which will migrate to the +ve charged plate) are stripped of electrons as they approach the cell boundary. Any negatively-charged ion will exhibit this behaviour in a strong enough potential well, but the OH⁻ ions have a strong tendency to such dissociation. This results, momentarily, in a region of negative-charge close to the positive cell boundary. Thus, on opposite sides of the active cell, there are hydrogen nuclei (free proton zone) and displaced electrons (-ve charge zone), both tending to increase in density closer to the charged plates.

If, at this point, the charge is removed from the plates, there is a tendency for the charge-zones to move, albeit very slowly, towards the centre of the active cell. The ion-transit rates of free electrons and of hydrogen nuclei are, however, some two orders of magnitude faster than either H₃O⁺ ions or OH⁻ ions.

If the charges are now replaced on the plates, but with opposite polarity, the interesting and potentially useful aspect of the process is revealed. Hydrogen nucleus migration is accelerated in the direction of the new -ve plate and free electron migration takes place towards the new +ve plate. Where there is a sufficient concentration of both species, including the accumulations due to previous polarity changes, monatomic hydrogen is formed with the liberation of some heat energy. Normal molecular association occurs and H₂ gas bubbles off from the cell.

Also existing OH⁻ radicals are further stripped of hydrogen nuclei and contribute to the process. O⁻ ions give up their electrons forming the diatomic molecule and similarly bubbling off from the cell.

- 7 -

Thus, the continuous application of a strong electric field, changing in polarity every cycle, is sufficient to disrupt water into its constituent gaseous elements, utilising a small fraction of the energy required in conventional electrolysis or chemical energetics, and yielding heat energy of the enthalpy of formation of the diatomic bonds in the hydrogen and oxygen.

Apparatus for performing the process described above is described below. In particular, electronic circuitry to effect the invention is shown in the simplified block diagram of Figure 1. In Figure 1 a pulse-repetition-frequency (PRF) generator 1 comprises an astable mvbr clock running at a frequency which is preset for any application, but able to be varied across a range of approximately 5-30 kHz. The generator 1 drives, by triggering with the trailing edge of its waveform, a pulse width (PW) timer 2. The output of the timer 2 is a train of regular pulses whose width is determined by the setting of the timer 2 and whose repetition frequency is set by the PRF generator 1.

A gate clock 3 comprises a simple 555-type circuit which produce a waveform (see Figure 3a) having a period of from 1-5 ms, e.g. 2 ms as shown in Figure 3a. The duty cycle of this waveform is variable from 50% to around 95%. The waveform is applied to one input of each of a pair of AND gates 5a, 5b and also to a binary divide-by-two counter 4. The output of the counter 4 is shown in Figure 3b.

The signal from the divide-by-two counter 4 is applied directly to the AND gate 5b serving phase-2 driver circuitry 7a but is inverted before application to the AND gate 5a serving phase-1 driver circuitry 7a. The output of the AND gate 5a is therefore ((CLOCK and (NOT (CLOCK)/2)) and the output of the AND gate 5b is ((CLOCK) and (CLOCK/2)), the waveforms, which are applied to pulse-train gates 6a and 6b, being shown at Figures 3c and 3d.

- 8 -

Trains of 5-30 kHz pulses are applied to drive amplifiers 7a and 7b alternately, with a small "off"-period during which no pulses are applied to either amplifier. The duration of each "off" period is dependent upon the original
5 duty cycle of the clock timer 3. The reason for the small "off" period in the driver waveforms is to prevent local corona arc as the phases change over each cycle.

The drive amplifiers 7a, 7b each use a BC182L transistor 10 (see Figure 2), small toroidal 2:1 pulse
10 transformer 11 and a BUZ11 power-MOSFET 12 and apply pulse-packets across primary windings of respective 25 kV line-output transformers 8a, 8b to produce an EHT ac voltage of high frequency at the secondary windings. The secondary windings are 'lifted' from system ground and provide, after
15 simple half-wave rectification, the applied field for application to cell 20 (see Figure 4).

The cell 20 comprises a container 21 having walls 21a, 21b of electrically insulating material, e.g. a thermoplastics material, such as polymethyl methacrylate,
20 typically spaced about 5 mm apart, and bipolar cell electrodes generally designated 22, 23 and typically constructed from aluminium foil, positioned outside the walls 21a and 21b. Each bipolar cell electrode comprises a pair of electrode plates 22a and 22b (or 23a and 23b) for
25 each side of the cell 20 separated from each other by an electrically insulating layer 24 (or 25), e.g. of polycarbonate plastics material about 0.3 mm in thickness. The electrode plates 22a and 23a form one set (set A) of electrode plates positioned on opposite sides of the
30 container 21 and the electrode plates 22b and 23b form another set of electrode plates positioned on opposite sides of the container 21. An insulating layer 25, e.g. of polycarbonate material, similar to the insulating layers 24a or 24b may be positioned between each bipolar cell electrode
35 22 (23) and its adjacent container wall 21a (21b). A liquid electrolyte 26, preferably water, is contained in the container 21.

- 9 -

In use a train of positive pulses is applied to the electrode plates 22a and 23b and a train of negative pulses is applied to the electrode plates 23a and 22b. The timing of the pulses is shown schematically in Figure 5, which illustrates that, for set A (or for set B), whenever a positive pulse is applied to electrode plate 22a (or 23b), a negative pulse is also applied to electrode plate 23a (or 22b). However the pulses applied to the electrode plate set A are "out of phase" with the pulses applied to the electrode plate set B. In each train of pulses, the duration of each pulse is shorter than the gap between successive pulses. By arranging for the pulses of electrode plate set B to be applied in the periods when no pulses are applied to the electrode plate set B, the situation arises where pairs of pulses are applied successively to the electrode plates of different sets of electrode plates, there being a short interval of time when no pulses are applied between each successive application of pulses to pairs of electrode plates. In other words, looking at Figure 5, pulses P1 and Q1 are applied at the same time to the electrode plates 22a and 23a. The pulses P1 and Q1 are of the same pulse length and, at the end of their duration, there is a short time period t before pulses R1 and S1 are applied to the electrode plates 23b and 22b. The pulses R1 and S1 are of the same pulse length as the pulses P1 and Q1 and, at the end of their duration, there is a further time t before the next pulses P2 and Q2 are applied to the electrode plates 22a and 23a. It will be appreciated that whenever a pulse of one sign is applied to one of the electrode plates of a set, a pulse of the opposite sign is applied to the other electrode plate of that set. Furthermore by switching from one to the other electrode plate set the polarities applied across the container are repeatedly switched resulting in an "alternating" electric field being created across the "liquid dielectric" water in the container.

- 10 -

CLAIMS

1. Fracture cell apparatus including a capacitive fracture cell (20) comprising a container (21) having walls (21a, 21b) made of non-electrically conducting material for
5 containing a liquid dielectric (26) and spaced apart electrode means (22, 23), and pulse applying means (1-8a, 8b) for applying pulses to each of the electrode means (22, 23),
characterised in that said spaced apart electrode means (22, 23) are positioned outside the container (21) with the
10 liquid dielectric (26) between the electrode means (22, 23) and in that the pulse applying means (1-8a, 8b) is for applying positive and negative pulses to each of the electrode means (22, 23) so that whenever one of a positive voltage pulse and a negative voltage pulse is applied to one
15 of the two electrode means (22, 23), the other of a positive voltage pulse and a negative voltage pulse is applied to the other of the two electrode means (22, 23), thereby creating an alternating electric field across the liquid dielectric (26) to cause fracture of the liquid dielectric.

20 2. Fracture cell apparatus according to claim 1, characterised in that the liquid dielectric (26) comprises water.

3. Fracture cell apparatus according to claim 1 or 2, characterised in that the pulse applying means (1-8a, 8b)
25 provides positive and negative step voltages at the same time to each electrode means (22, 23), the positive and negative step voltages being applied alternately to the two electrode means (22, 23) with a short period of time during each charge voltage cycle in which no step voltage is
30 applied to either electrode means.

4. Fracture cell apparatus according to any one of the preceding claims, characterised in that each electrode means (22, 23) comprises a bipolar electrode (22a, 22b; 23a, 23b).

- 11 -

5. Fracture cell apparatus according to claim 4, characterised in that each bipolar electrode comprises first and second plate means (22a and 22b; 23a and 23b), the first plate means of one bipolar electrode and the second plate means of the other bipolar electrode forming one electrode plate set and the second plate means of said one bipolar electrode and the first plate means of said other bipolar electrode forming another electrode plate set.

6. Fracture cell apparatus according to claim 5, characterised in that said pulse applying means (1-8a,8b) applies positive pulses to the first plate means of each of said bipolar electrodes and applies negative pulses to the second plate means of each of said bipolar electrodes.

7. A method of generating gas, characterised in that the method comprises applying positive and negative voltage pulses alternately to electrode means (22, 23) positioned either side of, but not in contact with, a liquid dielectric (26), the voltage pulses being applied so that, whenever one of a positive voltage pulse and a negative voltage pulse is applied to one of the two electrode means, the other of a positive voltage pulse and a negative voltage pulse is applied to the other of the two electrode means, the applied voltage pulses generating an alternating electric field across the liquid dielectric (26) causing fracture of the liquid dielectric into gaseous media.

8. A method according to claim 7, characterised in that voltage pulses of at least 15 kV, e.g. 25 kV, either side of a reference voltage are applied to the electrode means (22, 23) across the liquid dielectric (26) to generate the alternating electric field.

FIG. 1

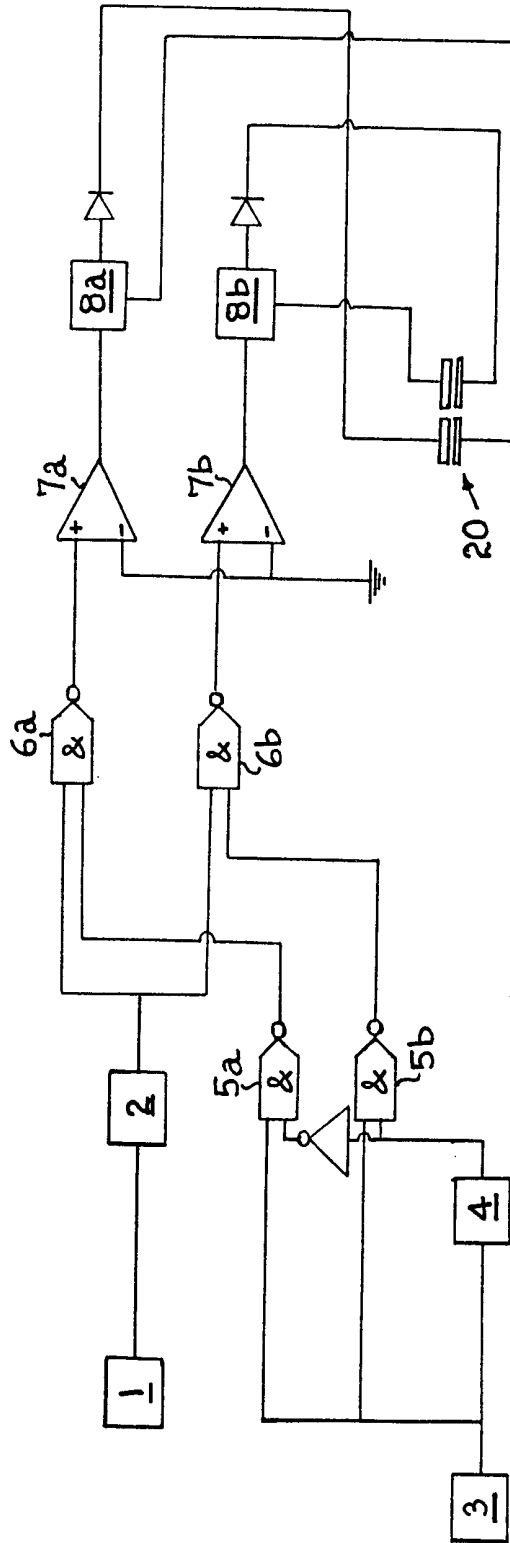


FIG. 2

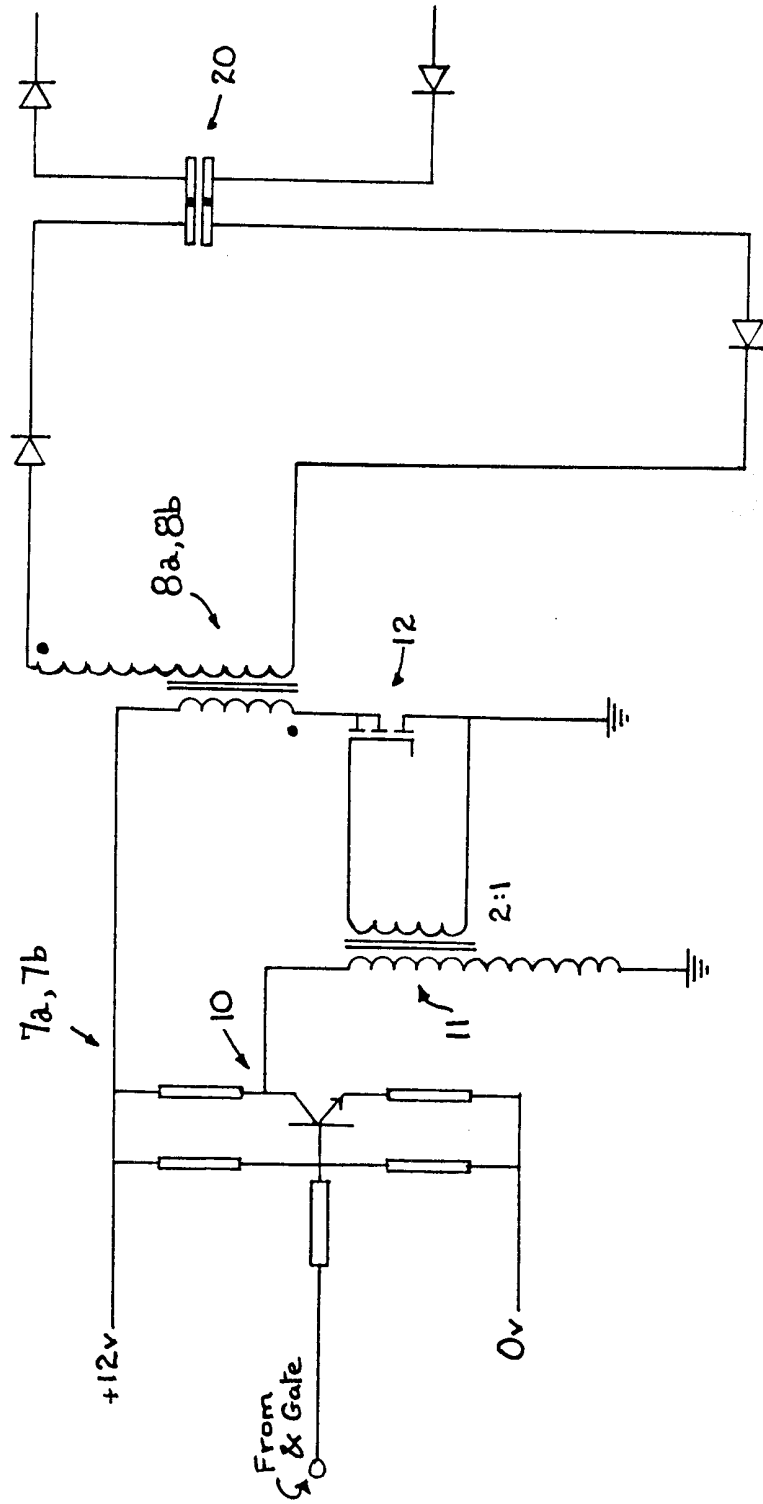


FIG. 3

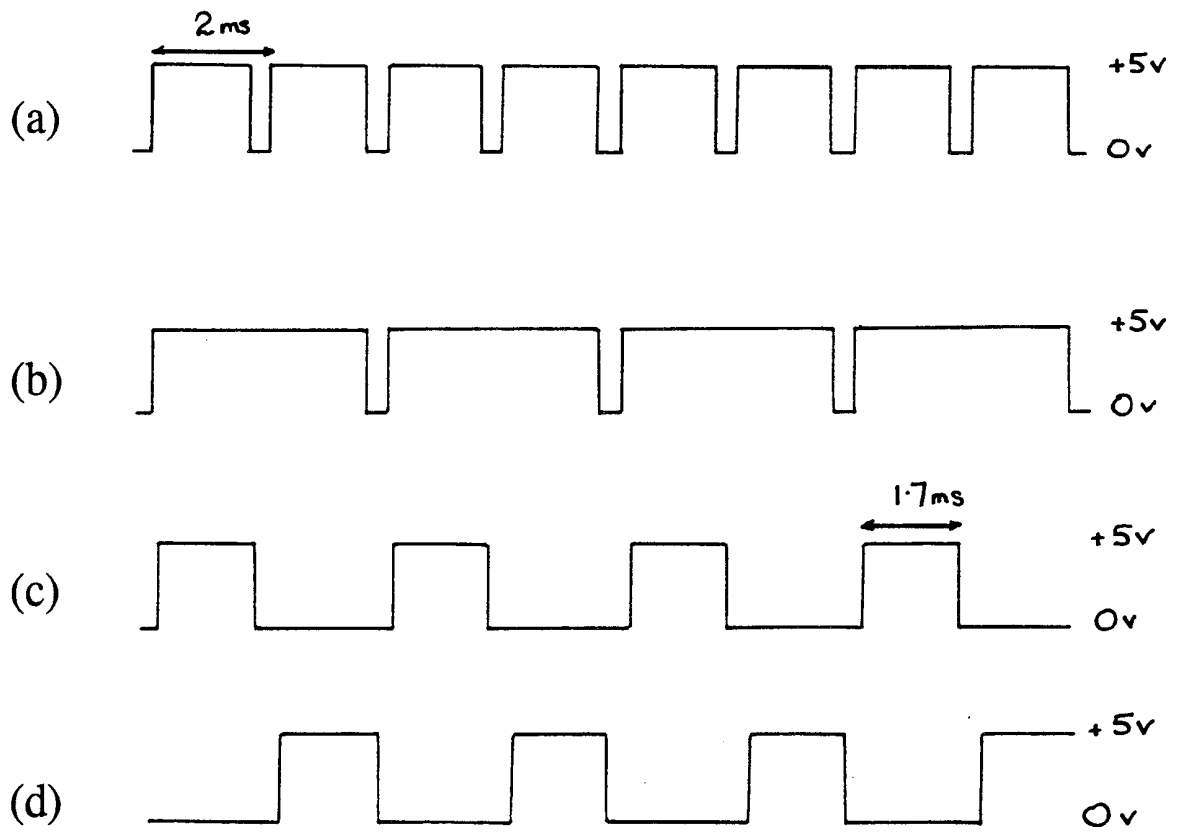


FIG. 4

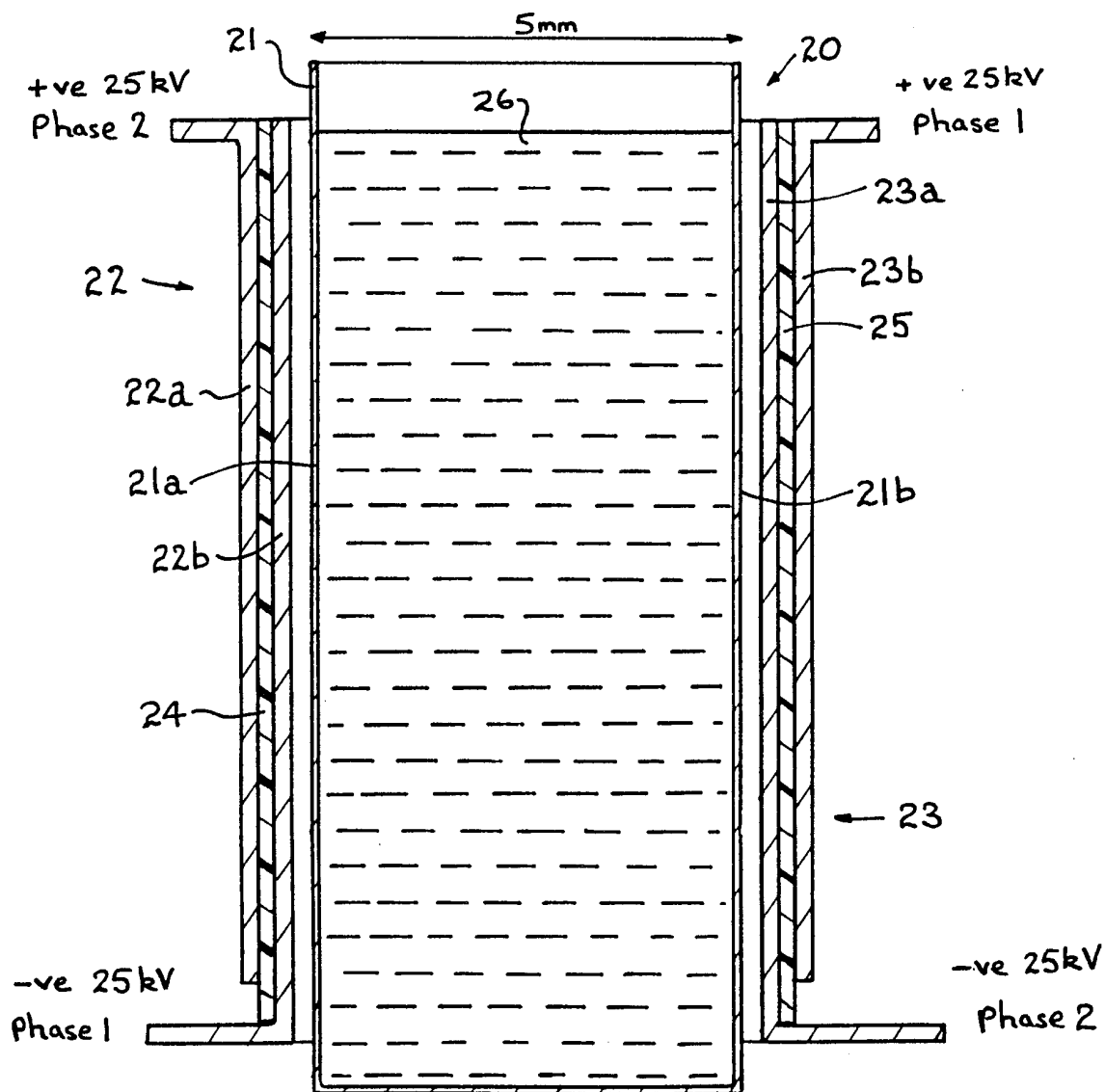
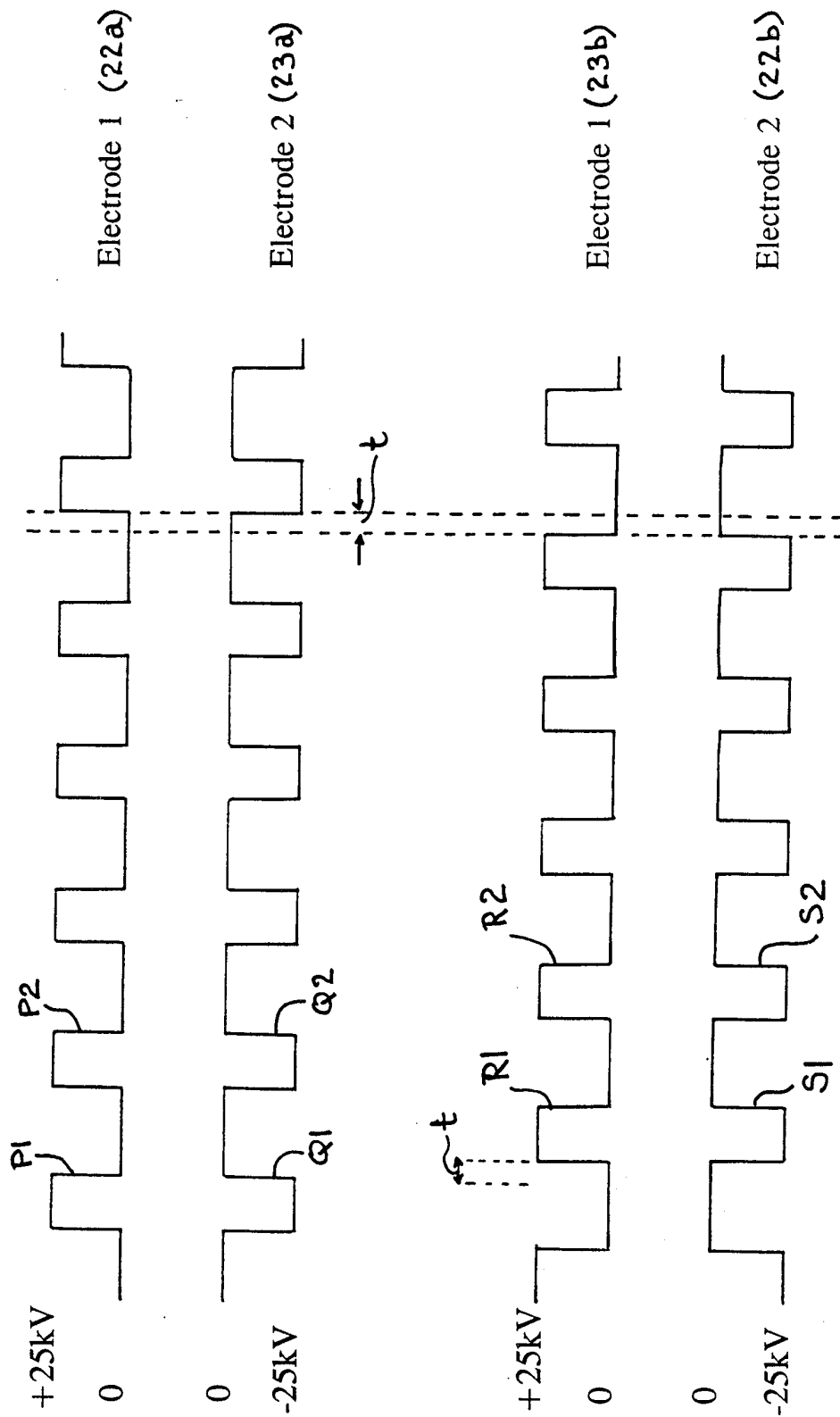


FIG. 5



INTERNATIONAL SEARCH REPORT

International Application No

PCT/GB 98/01099

A. CLASSIFICATION OF SUBJECT MATTER
 IPC 6 B01J19/08 C01B3/04 B01J7/02

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 B01J C01B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category ^o	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 4 427 512 A (TAY-HEE HAN) 24 January 1984 see the whole document ----	1, 2, 7, 8
A	GB 1 189 888 A (CHEMOLIMPEX MAGYAR VEGYIARU KULKERESKEDELMI VALLALAT) 29 April 1970 see the whole document ----	1, 3, 6, 7
A	WO 89 12704 A (STANLEY A. MEYER) 28 December 1989 -----	

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

^o Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

- "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- "&" document member of the same patent family

Date of the actual completion of the international search

18 August 1998

Date of mailing of the international search report

31/08/1998

Name and mailing address of the ISA
 European Patent Office, P.B. 5818 Patentlaan 2
 NL - 2280 HV Rijswijk
 Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
 Fax: (+31-70) 340-3016

Authorized officer

Stevnsborg, N

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/GB 98/01099

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 4427512 A	24-01-1984	NONE	
GB 1189888 A	29-04-1970	BE 698836 A CH 495772 A DE 1642793 A FR 1537725 A NL 6707205 A SE 337806 B US 3766050 A	03-11-1967 15-09-1970 02-03-1972 27-11-1967 23-08-1971 16-10-1973
WO 8912704 A	28-12-1989	US 4936961 A AU 3862389 A EP 0381722 A JP 3500042 T US 5149407 A	26-06-1990 12-01-1990 16-08-1990 10-01-1991 22-09-1992