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ЭЛЕКТРОЛИЗ ВОДЫ ПРИ ПОМОЩИ ИМПУЛЬСОВ ПОСТОЯННОГО ТОКА И ПЛАЗМЕННОГО РАЗРЯДА

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В работе проанализирован процесс импульсного электролиза и оценена его эффективность. Индуктивные импульсы высокого напряжения (до 1 мкс) появляются во второй катушке трансформатора после разрыва цепи в первой цепи и сразу направляются в электролизную ячейку, являющуюся частью второй цепи. Подтверждено, что при очень коротких импульсах напряжения ячейка ведет себя как заряжаемый конденсатор, образуя разницу напряжения между электродом и электролитом. После короткого импульса наблюдается длинный хвост разряда, во время которого происходит высокоэффективный (приближающийся к 100% эффективности) электролиз воды. Были сконструированы 4 и 5 электродные ячейки для изучения плазменного электролиза. Исследуя электролизные газы при помощи масс-спектрометра, было обнаружено, что при плазменном электролизе раствора K_2CO_3 количество выделяемого водорода можно увеличить, добавив мочевину $(NH_2)_2CO$. Это объясняется тем, что продукты разложения мочевины реагируют с кислородом, образуя газообразные оксиды CO_2 и NO_x и позволяя накапливать большее количество водорода.

Ключевые слова: индуктивные импульсы, импульсный электролиз, плазменный электролиз.

WATER ELECTROLYSIS WITH DC PULSES AND PLASMA DISCHARGE

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Pulse electrolysis process is analyzed and available efficiencies are estimated. The inductive high voltage pulses (up to 1 microsecond) induced in secondary transformer coil immediately after the circuit interruption in primary circuit, are applied to electrolysis cell in secondary circuit. It is confirmed that for very short voltage pulses the electrolysis cell behave as capacitor which is charged, and voltage gradient is formed on electrode/electrolyte interface. After the short voltage pulse a long discharge tail is observed, during which water is splitting into hydrogen and oxygen with efficiency close to 100%. Four-and five-electrode cell is constructed to study plasma electrolysis. Analyzing the composition of electrolysis gases with the mass spectrometer, it is found that low concentrations of hydrogen in plasma electrolysis of K_2CO_3 solutions, can be effectively enhanced by using urea $(NH_2)_2CO$ additive. Explanation is proposed that products of decomposition urea in solution attracts oxygen and forms CO_2 and NO_x gases, thus allowing the accumulation of hydrogen gas.

Keywords: inductive impulses, pulse electrolysis, plasma electrolysis.

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M. Vanags, J. Kleperis, G. Bajars, A. Lusis, Water electrolysis using electrodes with modified surface/volume. Journal of Physics: Conference Series 93 (2007) 012025.

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Main range of scientific interests: electronics, programming, renewable energy technologies

Introduction

At a time when fuel prices are rising only, and forecasts show that fossil fuel energy resources may soon come to an end, a crucial role is any possibility to save fuel and drive car economically. The internal combustion engine has low efficiency and more than half of fuel being burned is wasted into heat. The burning of fuel is resulting with exhausts and as it is shown by different researches [1, 2], the cars on streets are responsible for generation of 65-90% air polluting substances in large cities (St.-Petersburg, Riga as examples [1, 2]). One solution to help engine burn gasoline more efficiently and reduce exhausts is referred to "water-to-fuel systems" simply converting plain water into a gas called HHO (Hydroxy or Brown gas) [3-5]. This gas is then used as a supplement to normal gasoline or diesel fuel. Hydrogen holds significant promise as a supplemental fuel to improve the performance and emissions of spark ignited and compression ignited engines. Hydrogen as a fuel for automotive engines is generally coupled with advanced conversion systems (fuel cells) but, due to energy crises and environmental pollution, hydrogen fuelling of internal combustion engines is of great interest as well. Hydrogen has the ability to burn at extremely lean equivalence rations - seven times leaner than gasoline and five times leaner than methane (lean mixtures allow for complete combustion, decreasing carbon monoxide emissions) [5]. The flame velocity of hydrogen is much faster than other fuels allowing oxidation with less heat transfer to the surroundings what improves thermal efficiencies. The only drawback to hydrogen is that it is found nowhere as a gas on Earth, and even its lower heat value is greater than other hydrocarbon fuels it is less dense therefore a volume of hydrogen contains less energy [4, 5]. Therefore different on-board systems to produce hydrogen are elaborated worldwide, from which water electrolysers are most popular (see for example [3-5]). But the energy that is derived from the hydrogen-oxygen combustion can not be greater than the energy that is put into electrolyser to split the water. Typically it is smaller due un-efficiency of Faradic electrolysis process and not exceed 50% [6]. The voltage in practical devices of electrolysis is higher than thermo neutral cell voltage (1.48 V). Having such value of voltage some part of the electricity transforms into heat, which heats up the cell and requires additional cooling. The voltage, used in the process of electrolysis, is defined:

$$E = E_{atg} + loss, \quad (1)$$

where the *loss* is:

$$loss = E_{anode} + E_{cathode} + E_{mt} + IR, \quad (2)$$

where E_{anode} – activation overvoltage of the anode; $E_{cathode}$ – activation overvoltage of the cathode; E_{mt} – overvoltage of the mass transfer; IR – ohmic overvoltage (includes resistance in an electrolyte, on electrodes, leads).

This is main reason while research for effective electrolysis methods is very urgent. However the development of electrolysis devices is in progress nowadays as well along with the development of proton exchange membrane, which can be used in the water electrolysers and fuel cells, along with the development of high-temperature solid oxide electrolysers likewise the optimization of alkaline electrolysers. There are different ways of water splitting described in the literature that sharply differs from conventional water electrolysis [7]. The most common could be: thermo chemical, sonochemical, photocatalytic, biological water splitting; water splitting under the magnetic field and centrifugal force of rotation; pulse electrolysis and plasma electrolysis.

Pulse electrolysis – short summary. Ghoroghchian and Bockris [8] in 1956 already suggested that the pulse electrolysis is more effective than conventional electrolysis. Many new patents appeared in 1970ties [9–18] stating to be invented over effective electrolysis (i.e. the efficiency on current power is higher than 100%). The water splitting scheme described in patents initiated a huge interest, but nobody has succeeded in interpreting this scheme and its performance mechanisms up to now, and what is more important, nobody has succeeded in experimentally repeating devices described as well. There has been found only one study of water electrolysis, where inductive voltage pulse is used as a power supply [19]. The conclusion of this research is that this kind of water electrolysis efficiency is not dependant on the electrolysis power, thus being in contradiction to the conventional opinion of electrolysis. When inductive voltage pulse is delivered to water electrolysis cell, two different processes appears after pulse kinetics: rapid charge followed by slow discharge [20, 21]. It is possible to conclude form the pulse kinetics that the double-layer charging process has been separated from the electrochemical reaction, which has not been mentioned nor analysed in the previous researches [19-21].

Plasma electrolysis – short summary. Studying plasma electrolysis of liquid ammonia solution Hickling and Newn [22] noticed plasma ball and evolution of hydrazine (N_2H_4), which does not form during usual electrolysis of such solution. Volt-ampere characteristic of the plasma electrolysis divides into several different regions [22, 23] – Ohmical and non-persistent regions. The authors [23] consider that the Faraday's reaction is in process in this area as well as a process where the mechanism of gas emission is similar to the Hickling's radiolytic mechanism [22]. Mizuno et all [24, 25] and Jin et all [26] has found several anomalies of the plasma electrolysis near a cathode. Mizuno [24] observed four discreet regions of plasma electrolysis in his research about thermolysis or direct water pyrolysis in NaCl and Na_2SO_4 electrolytes. Depending on the conductivity of electrolyte the plasma flares up on the electrode easier if the conduction is higher.

The main objects of this research are pulse electrolysis and plasma electrolysis of water solutions.

Experimental part

The inductive voltage pulses were generated in the specific electric circuit (Fig. 1) allowing to apply high voltage short pulses to water electrolysis cell (the higher water resistance, the greater the amplitude of the pulse – Fig. 1).

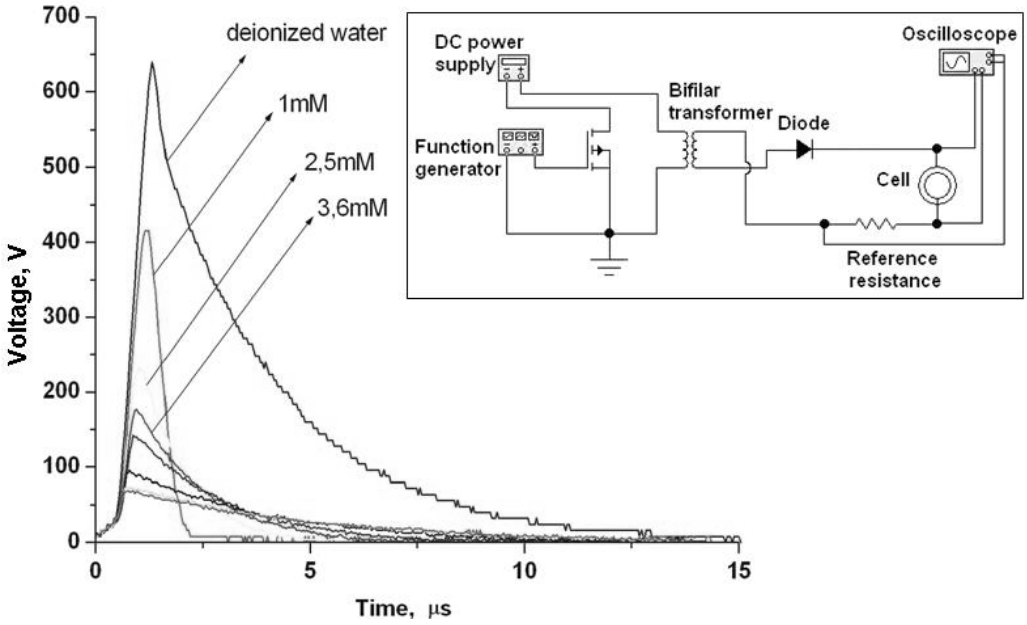

Рис. 1. Экспериментальная цепь для получения обратных индуктивных импульсов напряжения и записанные импульсы напряжения на электролизную ячейку, содержащую различные растворы KCl
Fig. 1. Experimental circuit for generation of inductive reverse voltage pulses and registered voltage pulses on electrolysis cell containing different water – KCl solutions

Circuit consist from of a pulse generator, a DC power source, a field transistor BUZ350, and a blocking diode A special broad-band transformer was bifilarly wound using two wires twisted together. Square pulses from the generator were applied to the field transistor connected in series with the DC power source. The filling factor of pulses was kept constant (50%). To obtain inductive reverse voltage pulses, the primary winding of the transformer is powered with low amplitude square voltage pulses. In the secondary winding (winding ratio1:1) due to collapse of the magnetic field induced in the coil very sharp inductive pulse with high amplitude and opposite polarity with respect to applied voltage appears. Pulse of induced reverse voltage is passed through the blocking diode, and the resulting ~1 μs wide high-voltage pulse is applied to the electrolytic cell (Fig. 1). A two-beam oscilloscope GWinstek GDS-2204 was employed to record the voltage (i.e. its drop on an reference resistance) and current in the circuit. Transistor IRF840 is used as semiconductor switch between DC power supply and grounding circuit. Pulse transformer is a solenoid type with bifilar windings; length is 20 cm and a coil diameter of 2.3 cm and ferrite rod core. Number of turns in both the primary and the secondary winding is 75, so ratio is 1:1. Inductance of solenoid is approximately 250 μH. Super-fast blocking diode with the closing time of 10ns is included in the secondary circuit, to pass on electrolysis cell only the pulses induced in transformer with opposite polarity.

Amount of released gases during electrolysis was determined with volume displacement method. Specific electrolysis cell was made for study the kinetics of inductive pulse electrolysis in very diluted electrolytes, consisting from glass bowl with two separate electrode holders equipped with screws for electrodes from stainless steel 316L wires (diameter 2 mm), length 100 mm).

Plasma electrolysis cell was constructed to observe arc discharge between two electrodes, one from which is slightly immersed tungsten wire (diameter 2 mm), second – large area steel plate electrode with area 24 cm^2 (Fig. 2). Embedded in the cell is independent electrode pair - two stainless steel electrodes with equal areas (4 cm^2) to register the changes in electrolyte during plasma discharge process.

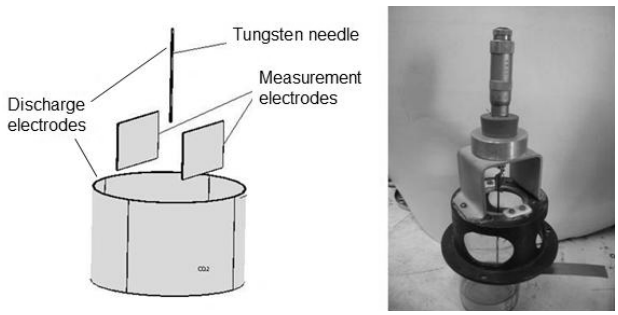

Рис. 2. Ячейка для плазменного электролиза с частично погруженным вольфрамовым проволочным электродом
Fig. 2. Plasma electrolysis cell with slightly immersed tungsten wire electrode

The tip of slightly immersed wire may be considered as paraboloidal shape electrode; immersion depth is changed by a micrometer screw from 0.5 to 1.5 mm. Water solution of 0.2 M K_2CO_3 with urea addition was used as electrolyte. Mass-spectrometer RGAPro100 was used to measure gas composition during plasma electrolysis processes.

Results and discussion: short pulse electrolysis

Current changes the direction from negative to positive with increasing concentration of electrolyte passing through the point where the current pulse has not descending a long tail (Fig. 3).

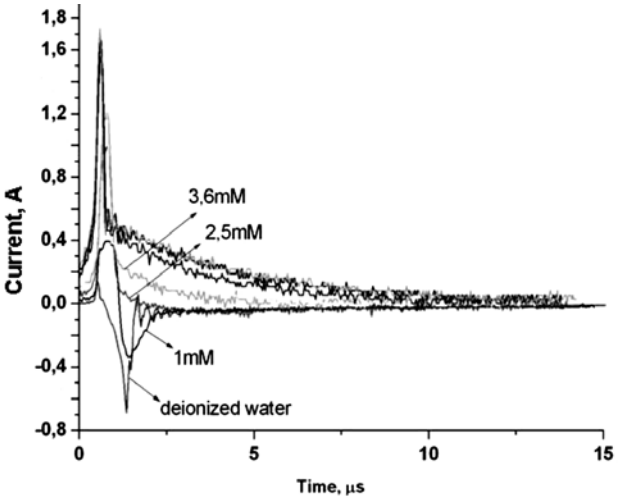

Рис. 3. Импульсы тока, возбужденные индуктивными импульсами напряжения в ячейке с различными растворами КОН
Fig. 3. Current pulses initiated by inductive voltage pulses on cell with different concentrations of KOH solution

Current pulse in deionised water most of the momentum is negative. By increasing the concentration of solution up to 1 mM, current pulse appears in both positive and immediately following a negative pulse, while discharge tail almost disappears. Continuing to increase the concentration of electrolyte the negative values of current pulse disappear and the discharge tail remains positive and increasing, which indicates that the charge injected in the cell during pulse increases. More increase of concentration does not change the view of current pulse and it remains like from the previous concentrations (Fig. 3).

When looking at pulse generation scheme and amplitudes of pulses applied o electrolysis cell (Fig. 1), it is clear that high-voltage pulse generated in the transformer is reactive in nature. Reactive pulse amplitude will depend on the quality factor of capacitive element. Capacitor with a large leak (concentrated electrolyte solution) will not be able to hold the reactive pulse with large amplitude, but it is always greater than the direct pulse amplitude. This means that at the first

moment when short inductive pulse is applied, the water electrolysis cell behaves as good capacitor, also at the voltage region, in which water electrolysis can occur. But after starting the discharge tail, the energy stored in the capacity transforms into the chemical energy in the process of water electrolysis.

Concerning occurrence of the negative currents following hypothesis is proposed. Voltage pulse kinetics demonstrates that around the electrode spatial charge density appears, i.e., when voltage rapidly grows in two-electrode system, electrons are emitted from the cathode environment (Fig. 4).

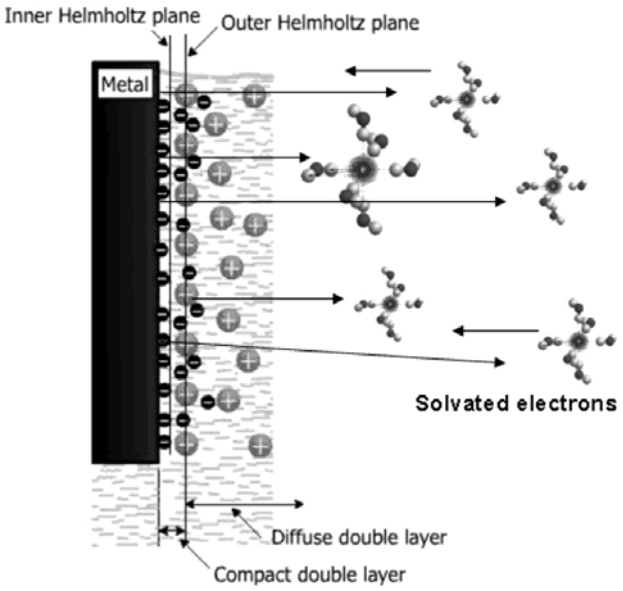

Рис. 4. Схема границы электрода и электролита с двойным слоем и испущенными/сольватированными электронами
Fig. 4. The scheme of electrode/electrolyte interface with double layer and emitted/solvated electrons

Since water ion concentrations in deionized water is low (H_3O^+ molar concentration is in the order of 10^{-7} M), then, most likely, the emitted electrons are solvated between polar water molecules and than will attach a neutral water molecule, which is described by following hydratization reaction:

$$H_2O + e^- \rightarrow H + OH^-. \qquad (3)$$

If OH^- ions and solvated electrons don't manage to discharge at the cathode, then around the cathode a spatial charge appears. In case of arising the spatial charge around electrode, it is more likely of electrons to move back into the metal. If the electron donor is the OH^- ion, then oxygen evolution should appear at the cathode. In principle, according to the experimental circuit, such electron returning back in metal in large amounts what results from the negative current pulse value presented in Figure 1, is not possible since this current component is blocked by the diode incorporated in the circuit. Therefore behind the diode, parasitic element with the inductive nature must exist in the measurement circuit (Fig. 1) which becomes comparatively small and solvated electrons are discharged by ions in electrolyte, therefore decreasing negative current.

To confirm this hypothesis, it is necessary to determine if oxygen does not appear near the cathode (the solvated electrons OH^- form allows a reverse reaction (3) on the cathode). The concentration of dissolved oxygen in a solution near the cathode during pulse electrolysis in dependence of time was measured with oxygen microsensor [27]. During the first 60 seconds current pulses has an explicit negative tail in the cell. After 60 seconds, the generator is set to manage the negative tail disappearance. When an anodic current tail occurs, the oxygen evolves at the cathode, but when the anodic current during voltage pulses is prevented, the oxygen at cathode is no longer released.

Results and discussion: plasma electrolysis

Influence of plasma discharge on properties of solution was done with repetitive scanning of potential between two measurement electrodes in cell (Fig. 2). As it is seen from corresponding volt-amperic curves (Fig. 5), electrolyte environment between measuring electrodes are all affected when the plasma discharge ball appears on the tip of tungsten wire.

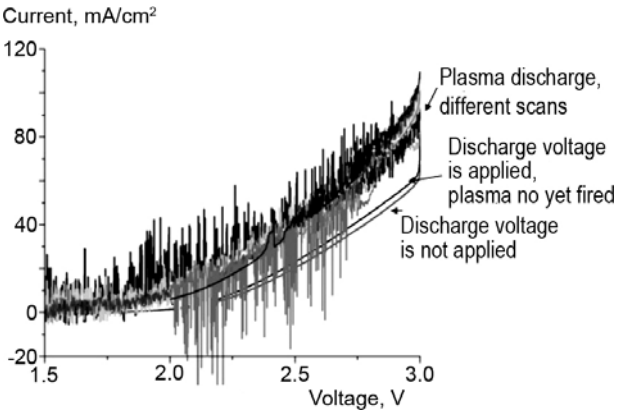

Рис. 5. Вольт-амперные характеристики для измеренных электродов в плазменной ячейке для электролиза с частично погруженной вольфрамовой иголкой
Fig. 5. Voltamperic characteristics of measuring electrodes in the plasma electrolysis cell with slightly immersed tungsten needle

Important observation is that the electrolysis process which takes place between the measuring electrodes becomes more and more intense in the presence of plasma discharge – hydrogen evolution potential of the electrolysis reaction is reduced by 500 mV, and slope of VA curve in hydrogen evolution region increases. The observed results can be explained with two different reasons: whether there is an electromagnetic link between two independently connected systems, or whether plasma discharge affects conductivity of

electrolyte. If the first reason is excluded, then the changes of electrolyte properties could be explained, for example, with the temperature increase. In result of the plasma discharge the temperature of environment increases, with the result that increase ion mobility and electrolyte conductivity accordingly.

The presence of additional thermal energy from plasma discharge accelerates the beginning of the activation process of electrolysis on measuring electrodes. Not excluded also the ionization of the electrolyte, it is - an increase of its conductivity. Slightly immersed tungsten wire electrode, fitted with a high voltage, emitted electrons into the environment, therefore an inflow of electrons in the electrolyte environment creates an imbalanced charge distribution, which is compensated by additional ion origination in the environment. Hence ion concentration in the environment increases, that causes the conductivity increase of electrolyte. If the current plateau in the plasma electrode circuit occurs due to the formation of thin isolating gas layer on the tip of tungsten wire, then, while it is in the plateau region, the tip of the wire has to cool down because current intensity decreases in the circuit. This means that the plateau is unstable and, if the voltage is not continued to increase in the wire's circuit, then current in the measuring cell's circuit would decrease. At plasma ignition, a current peak should appear at the plasma electrode circuit. When in the moment of ignition anomalous electron emission has started in the environment, then second plasma electrode which is deeply immersed has to compensate the excess charge and current has to increase.

Состав газов при плазменном электролизе с частично погруженным вольфрамовым проволочным электродом в 3% K_2CO_3 без и с добавлением мочевины (концентрация газа в объемных %, относительная ошибка 0,1%)

Gas composition in plasma electrolysis with slightly immersed tungsten wire in 3% K_2CO_3 without and with urea addition (gas concentrations in volume %, relative error 0.1%)

Gas	Reference spectrum, when cell is filled with argon	Plasma discharge in 3% K_2CO_3	Plasma discharge in urea – water solution with 3% K_2CO_3
Ar	80.7	81.1	90.2
CO_2	< 0.1	< 0.1	0.7
H_2	< 0.1	0.9	4
NO	< 0.1	< 0.1	0.1
N_2	14.7	10.4	3.1
O_2	3.4	6.6	0.8
H_2O	0.9	0.9	1

Mass spectrum of plasma gases evolved by electrolysis with slightly immersed tungsten wire electrode is collected in Table. Before analysis the camera of electrolysis cell was filled with argon. Gas sample for masspectrometer analyses was taken with syringe. Gas samples were taken before plasma ignition and after ignition when plasma is stabilized for 1 minute.

Analyzing the composition of electrolysis gases with the mass spectrometer, it is found that low concentrations of hydrogen in plasma electrolysis of K_2CO_3 solutions, can be effectively enhanced by using urea $(NH_2)_2CO$ additive. Explanation is proposed that products of decomposition urea in solution attracts oxygen and forms CO_2 and NO_x gases, thus allowing the accumulation of hydrogen gas.

Water plasma discharge with graphite electrodes is suitable for combustible gas (hydrogen + oxygen + carbon monoxide) production. After experimental work it was established that graphite electrode quickly burned out, causing the CO gas and carbon nano-sized particles, which converts electrolyte into a coal emulsion and catalyzes the subsequent discharge process. Hence, to use such plasma discharge in practice, a device is necessary for a continuous feed of graphite electrode in a plasma discharge zone.

Conclusions

Reactive short voltage pulse generator is designed to power water electrolysis cells of different constructions, both with spatially separated and with variable distance electrodes. Required value of electrolysis voltage in the primary circuit of power supply can be reduced by inserting the electrolysis cell in secondary circuit of power supply together with inductive element and reverse diode.

By changing the distance between the electrodes and concentration of electrolyte, it is experimentally proved that the electrolysis cell is capacitor with high Q factor when short voltage pulse (width below 1 μs) is applied. During this short time the capacitor (electrolysis cell) is charged, which can be interpreted as charging of double-layer on interface cathode/electrolyte. After the interruption of short voltage pulse the energy accumulated in double-layer capacitor slowly discharges (pulse discharge tail), thus activating the process of electrolysis. Consequently, it is shown that with short voltage pulse electrolysis the charging of cell can be separated from the electrochemical reactions in electrolysis process.

There has been discovered that the urea mixed to 0.2 M K_2CO_3 water solution markedly increases the amount of burning gas (hydrogen, carbon oxides, hydrocarbons and nitrogen oxides NO_x) in the process of plasma electrolysis with tungsten electrodes, with more than 4% by volume. This suggests that plasma electrolysis decomposes the urea and released gases attract oxygen, allowing the increase of hydrogen concentration in the surrounding space.

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http://www.hhoxygen.com/Default.asp?Redirected=Y;
http://waterpoweredcar.com/hydrobooster.html;
http://www.cargaswater.com/;
http://www.fuelfromh2o.com/;
http://diyhydrogenhho.com/;
http://www.orb.lv/blog/?item=98689;
http://waterfuelforall.com etc.

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