

SEPARATION BETWEEN CHARGING AND CHARGE TRANSITION CURRENTS WITH INDUCTIVE VOLTAGE PULSES

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Abstract

Inductive voltage pulses are generated in electric circuit consisting from DC power source, pulse generator, field transistor BUZ350, blocking diode and transformer wound as bifilar. Very short inductive voltage pulses arising from current disruption in primary circuit ($>1 \mu\text{s}$) are applied to water electrolysis cell. Quick cell charging is observed, which transforms in a relatively slower discharge tail. The oscilloscope is used to take voltage and current pulses from the cell consisting from steel electrodes and water-KOH solution. By changing the concentration of electrolyte and distance between electrodes, it is proved that using inductive voltage pulses to power electrolysis cell, it is possible to separate both the double layer charging current from the charge transition (Faradic) current.

Key words: water electrolysis cell, hydrogen evolution reaction, inductive reverse voltage pulse, high voltage pulse electrolysis

Introduction

Water electrolysis history started with observations of Nicholson and Carlisle in 1800 [1]. There were around 400 industrial water electrolysis units on 1902 and first large water electrolysis plant with production capacity $10000 \text{ Nm}^3/\text{h}$ was opened on 1939 [2]. High pressure electrolyzers appeared some years later, but first industrial solid polymer electrolysis system was made by General Electric on 1966. High temperature solid oxide electrolyzers were introduced in 1972, but first large alkaline electrolysis unit was built on 1978 [2]. Although the development of different water electrolysis systems are evident by walking through the time, their

energy efficiency is still under 60% in operational mode, calculating on lower hydrogen combustion heat [3,4]. Here the energy efficiency means relation between combustion heat obtained from evolved hydrogen mass to the electricity consumed by electrolysis. So far, the world's main research direction is the high-temperature electrolysis and polymer electrolyte membrane (PEM) electrolyzers [3]. With high temperature electrolysis the increase of efficiency is achieved making the heat input to activate water splitting. Nevertheless, the high temperature system requires power to be supplied as heat, which often is not easy. On the other hand the PEM electrolyzers are working under normal conditions with a relatively satisfactory efficiency - around 60%, which is still lower the required efficiency of energy storage scheme, furthermore the PEM electrolysis systems are more expensive than alkaline electrolyzers nowadays [4].

Typically direct current (DC) power is used in electrolysis; nevertheless pulse DC voltage also can be used [5–7]. Bockris et al [6] found that applying voltage pulses on an electrolysis cell, the long current tail is observed just after end of voltage pulse. Shimizu et al [5] used inductive voltage pulses (200 ns) to power electrolysis cell and found that efficiency of electrolysis does not change with changing applied power. We also used inductive voltage pulses [7] to compare different metals as cathode and found that concentration of dissolved hydrogen grows faster on metals with higher hydrogen evolution overvoltage and lower hydrogen solubility.

In this work we prove the fact that using inductive voltage pulses to power electrolysis cell, it is possible effectively reduce applied potential thanks to the possibility to separate both the charging current from the charge transition (Faradic) current in hydrogen evolution reaction.

Experimental setup

Self-made electrolysis cell was used in experiments (Figure 1). It is made from poly-acetal body with two stainless steel SUS316L electrodes. One electrode is fixed but second is moving and connected to micro-screw. Special sealing holes are made to avoid micro-screw holder material to contact electrolyte. Before experiments sealing holes are filled with isolating mastic. Electrochemical cell is in a form of hollow with diameter 40 mm, but steel electrodes are squares with area 2 cm². Before experiments

electrodes were mechanically polished and washed with acetone and deionised water. As electrolyte the KOH solution with concentrations 0,1 – 0,5 M are used. During experiments the distance between electrodes was varied from 1 to 5 mm.

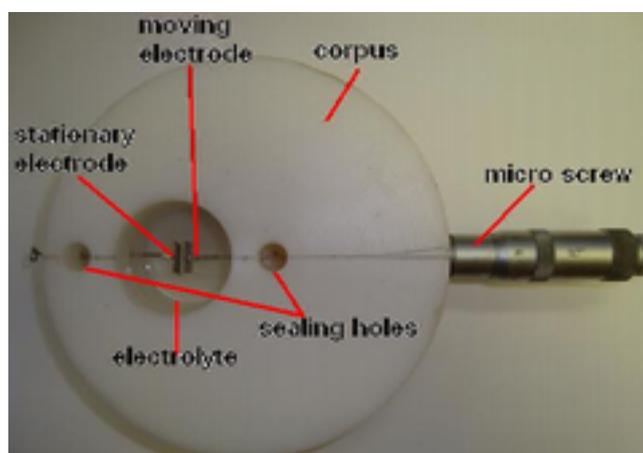


Figure 1. Water electrolysis cell.

Inductive voltage pulses were generated with an electric circuit (Figure 2) consisting from pulse generator, DC power source, field transistor BUZ350 and blocking diode [7,8]. Special transformer is wound as bifilar from two wires twisted together. Square pulses from generator are used to conduct field transistor, connected in series with DC power source. The filling factor of pulses was kept constant – 50%. Obtained inductive reverse voltage pulses powers the primary winding in bifilar transformer. After each square pulse in this broad band bifilar transformer the very sharp and high inductive pulse with opposite polarity is induced in secondary winding of transformer (winding ratio 1:1) due to the collapse of magnetic field induced in the coil. Pulses of induced reverse voltage are directed through blocking diode and resulting high voltage pulses with width around 1 μ s are obtained and used to power electrolysis cell. Two beam oscilloscope GWinstek GDS-2204 is used to register voltage and current (voltage drop on an etalon resistance) in circuit powering electrolysis cell.

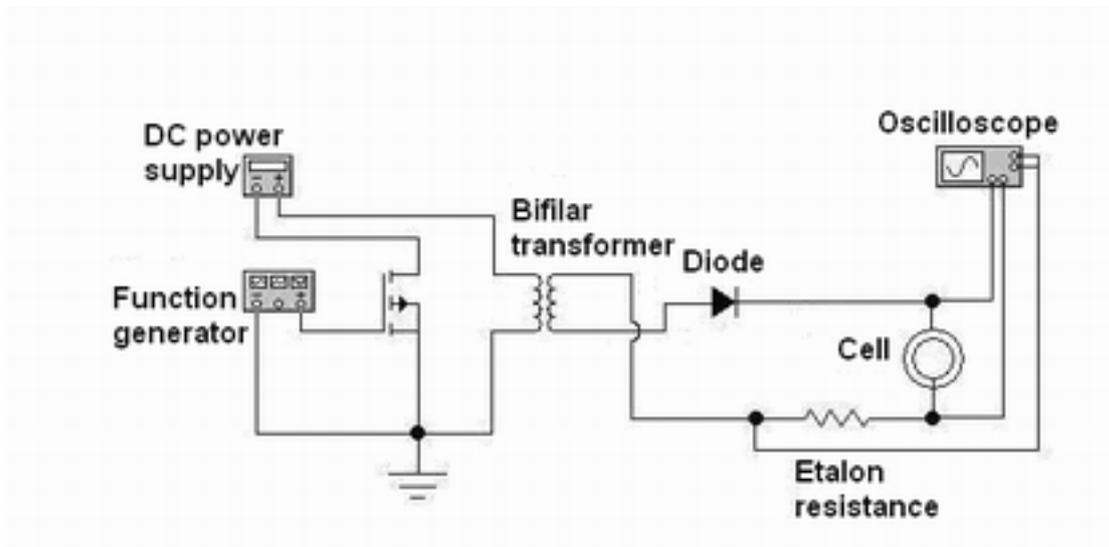


Figure 2. Experimental circuit for generation of inductive reverse voltage pulses.

Results and discussion

After applying inductive voltage pulse to water electrolysis cell, a typical oscillogram appears on the oscilloscope (Figure 3). Oscillogram is taken in 3M KOH solution, while the electrodes are 3 mm away from each other. In all experiments the pulse repetition time was chosen equal to 50 microseconds and direct pulse amplitude was set at 1 V. Both the voltage pulse and current pulse are with fast rising front. Voltage grows up to a maximum (amplitude depends on the distance between the electrodes), followed by a long decreasing tail. When the end of tail is reached, the current is approaching 0, but the voltage stays around value 2 V. Next pulse is identical to previous and so on.

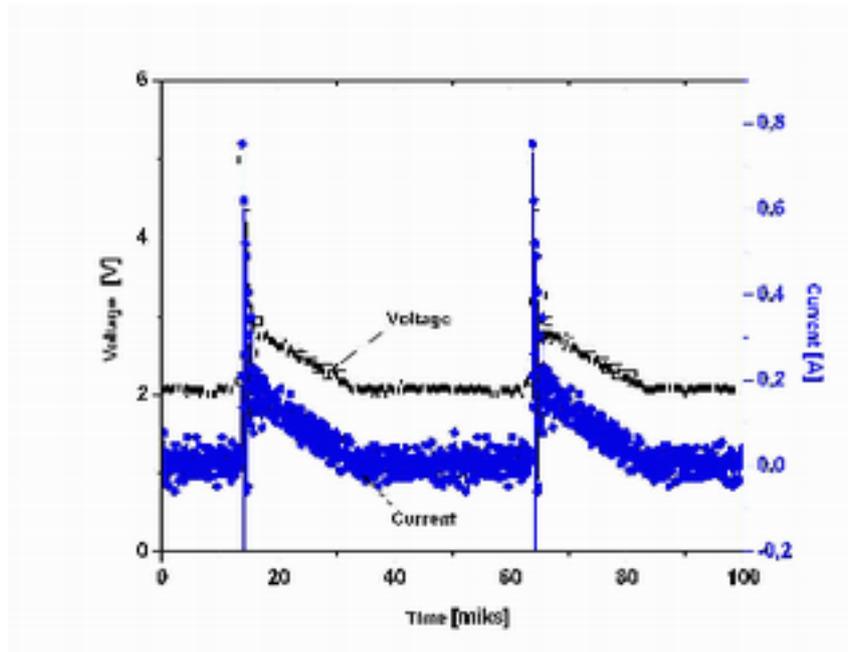


Figure 3. Typical voltage and current pulses on the screen of oscilloscope when inductive reverse voltage pulses are applied to electrolysis cell.

The voltage and current oscillograms in 0.1, 0.3 and 0.5 M KOH solutions respectively (Figures 4-6) were measured for different distances between electrodes. As it is seen in 0.1 M KOH solution, the maximum induced voltage pulse value is about 5.5 V at the distance 5 mm between electrodes, and only 3 V when distance between electrodes is 1 mm. In solution with higher concentration (0.3 M KOH) the voltage pulse reaches only 3,5 V at the distance between electrodes 5 mm, and drops to 2.6 V when the distance between electrodes is reduced to 1 mm. More concentrated solution, ie, 0.5 M KOH, characterizes with more lower voltage pulse values – 2,9 V at the distance 5 mm and 2,4 V at the distance between electrodes 1 mm. The peak values of current does not change significantly in dependence on the electrode distance and solution concentration, but the length of discharge tail is changing, suggesting that the larger charge amount is transmitted in most concentrated solution.

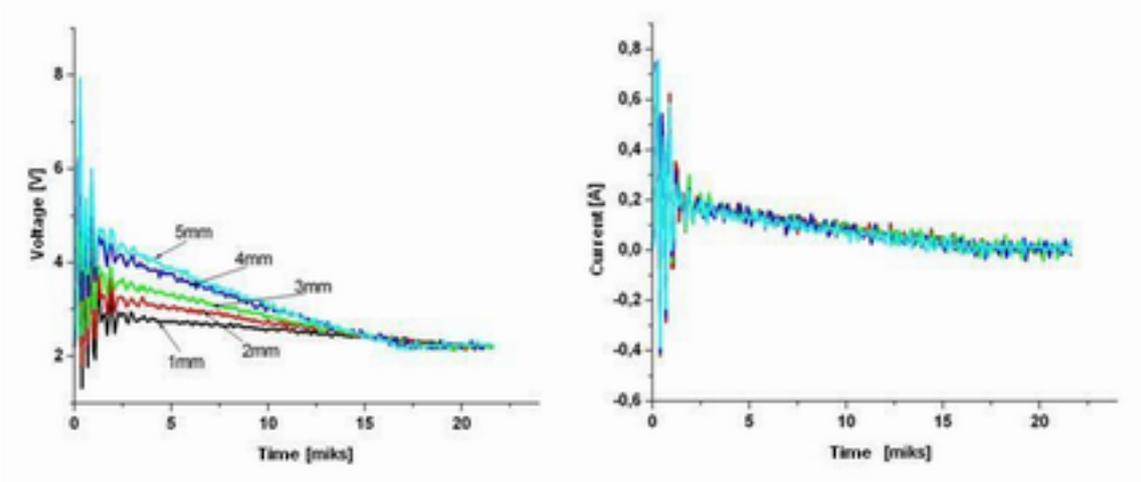


Figure 4. Voltage and current oscillograms in 0.1 M KOH solution

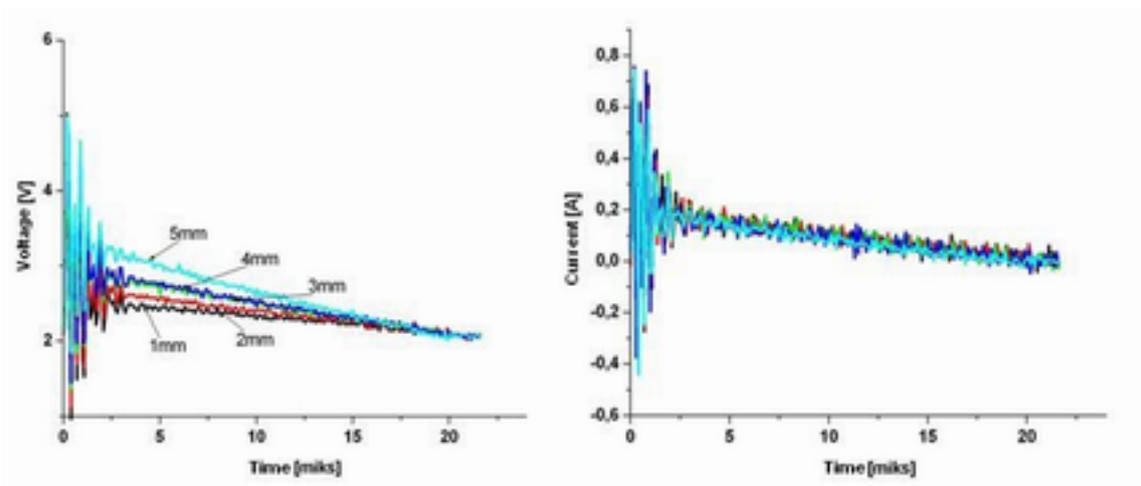


Figure 5. Voltage and current oscillograms in 0.3 M KOH solution

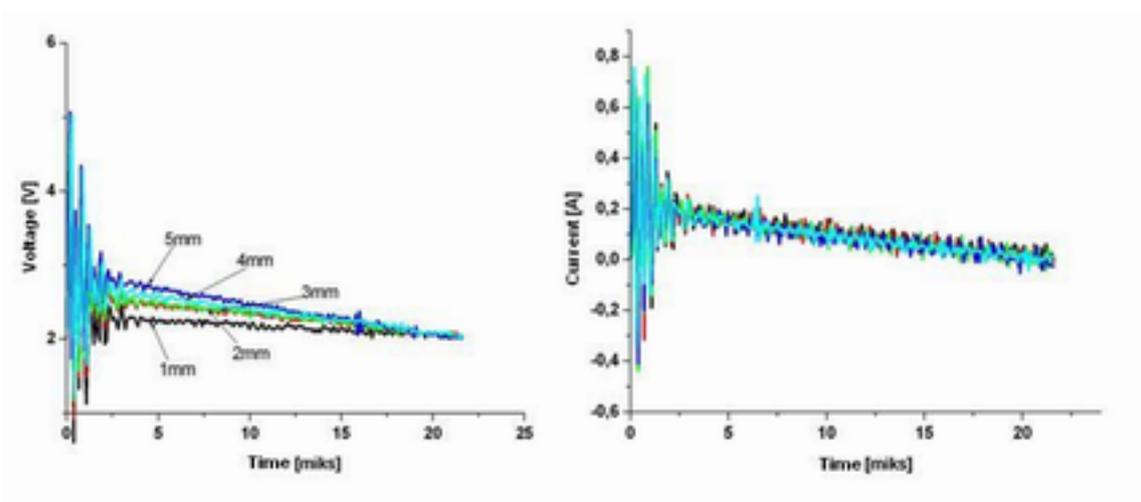


Figure 6. Voltage and current oscillograms in 0.5 M KOH solution

Looking at the pulse generation scheme in Figure 2, it is clear that the nature of resulting high voltage pulse created in transformer is reactive. This is because the amplitude of an active pulse in circuit's primary side does not exceed value 1 V, but on secondary side voltage exceeds value 2 V and is reversed due blocking diode in circuit. Reactive pulse amplitude depends from the quality factor (Q) of the capacitive element. Capacitor with a large leak can not keep high amplitude for inductive voltage pulse, but from Figures 3-6 above it is seen that the reverse pulse amplitude is achieved higher as amplitude of direct pulse.

Typical equivalent scheme of an electrochemical cell consists from parallel circuit of capacitor and resistor connected in serial with another parallel circuit of capacitor and resistor. The first parallel circuit reflects the geometrical capacity of the cell and resistivity from oriented molecules, while the second parallel circuit represent the capacity of double layer on an interface electrode/electrolyte and resistivity from Faraday hydrogen evolution reaction. Due to the fact that the geometric capacity compared with the double layer capacity is much smaller, then the first parallel circuit make little contribution to the overall picture.

When voltage pulse is applied to the electrochemical cell, the double layer capacity is charged and this charging current is registered. In parallel, when the voltage on double layer between the electrode and solution exceeds the value of red-ox normal potential, the circumstances will be provided when Faraday current will appear. The oscillograms in Figures 3-6 gives evidence that the Faraday current has not time to appear in the first microsecond and thus an amplitude of reactive pulse is increasing. In other words, the cell behaves like a capacitor with great Q in the first microsecond. The more concentrated the solution, the higher capacity of double layer capacitor, therefore at the same value of charge the voltage should be reduced – compare Figures 4-6. By reducing the distance between the electrodes, double layer capacity is growing even faster, because the maximal value of voltage peak decreases significantly, when the distance between electrodes is reduced.

The kinetics of voltage and current pulses changes significantly immediately after charging the double layer capacitor, switching to steady discharge tail. Blocking diode does not allow the capacity to discharge in any other way than Faraday reaction until existing potential is unable to activate the electrochemical red-ox process (the value of existing potential drops below the value of red - ox normal potential).

It is concluded by Shimizu et al [6] that using an active voltage pulse in water electrolysis cell, the charging takes up too much of a pulse-time tracking. That fact does not favour a pulsed electrolysis comparing with the conventional DC electrolysis. But if a reactive inductive voltage pulse is used, previously mentioned lack is transformed into an advantage because after such a short charge period the discharge tail appears several times longer than that of the pulse length taken to infinite resistance.

Conclusions

Using to water electrolysis the very short voltage pulses arising from current disruption in primary circuit, quick cell charging is observed in secondary inductive circuit, which transforms in a relatively slower discharge tail. Authors believe that such kinetics can occur when electrochemical charging process of the cell is separated from Faraday charge transfer process. The water electrolysis cell behaves as a capacity with great quality factor when sharp pulse is applied, while the Faraday discharge takes place in the discharge tail, when power stored during short pulse charging is used.

Proposed inductive voltage pulse power scheme allows reducing the power in the side of the primary circuit, thereby to reduce the total power necessary for electrolysis. It is demonstrated that electrolysis of water and water-alkali solutions can be achieved with input voltage only 1 V. Great advantage of proposed pulse electrolysis is the fact that the charging of electrolysis cell lasts for a relatively shorter period than following discharge using previously stored energy.

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UZLĀDES UN LĀDIŅA PĀRNESES STRĀVU ATDALĪŠANA ŪDENS ELEKTROLĪZĒ AR INDUKTĪVIEM SPRIEGUMA IMPULSIEM

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Tēzes

Induktīvi sprieguma impulsi ar garumu $>1 \mu\text{s}$, kas tiek ģenerēti ķēdē, ko veido līdzstrāvas barošanas avots, impulsu ģenerators, lauka tranzistors BUZ350, bloķējošā diode un bifilāri tīts transformators, tiek izmantoti ūdens elektrolīzei šūnā, ko veido nerūsējošā tērauda elektrodi un ūdens – sārma šķīdums. Oscilogrāfs tiek izmantots, lai reģistrētu sprieguma un strāvas impulsus elektrolīzes šūnā. Novērots, ka šūnas uzlāde notiek ļoti strauji, kam seko lēni krītoša izlādes aste. Mainot elektrolīta koncentrāciju un attālumus starp elektrodiem, pierādīts, ka ātrā induktīvā sprieguma impulsa laikā elektrolīzes šūna uzvedas kā kondensators, kas tiek uzlādēts. Uzlādē uzkrātā enerģija pēc tam tiek izmantota elektrolīzes reakcijai, kas atspoguļojas lēni krītošā strāvas un sprieguma astē. Tādējādi tiek atdalīti dubultslāņa uzlādes un Faradeja elektrolīzes procesi, kas ļauj realizēt ūdens elektrolīzi ar nelielu enerģijas patēriņu.

Atslēgas vārdi: ūdens elektrolīzes šūna, ūdeņraža izdalīšanās reakcija, induktīvs sprieguma impulss, augstsprieguma impulsu elektrolīze.