

SEPARATION OF CHARGING AND CHARGE TRANSITION CURRENTS
WITH INDUCTIVE VOLTAGE PULSES

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Inductive voltage pulses are generated in the electric circuit consisting of a DC power source, a pulse generator, a BUZ350 field transistor, a blocking diode, and a bifilarly wound transformer. Very short inductive voltage pulses arising at disruption of current in the primary circuit ($>1 \mu\text{s}$) are applied to a water electrolysis cell, which causes its quick charging followed by a relatively slower discharge tail. To take voltage and current pulses from the cell consisting of steel electrodes and water-KOH solution, an oscilloscope is employed. By changing the concentration of electrolyte and the distance between electrodes it is found that applying inductive voltage pulses to such a cell it is possible to separate the double-layer charging currents from the charge transition (Faradic) current.

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

1. INTRODUCTION

The water electrolysis history started with observations of Nicholson and Carlisle in 1800 [1]. By 1902 about 400 industrial water electrolysis units could be counted in the world, and the first large water electrolysis plant with a production capacity of $10000 \text{ Nm}^3/\text{h}$ was opened in 1939 [2]. High-pressure electrolyzers appeared some years later, while a first industrial solid polymer electrolysis system was made by General Electric in 1966. High-temperature solid oxide electrolyzers were introduced in 1972, whereas a first large alkaline electrolysis unit was built in 1978 [2]. Although the development of different water electrolysis systems is evident when walking through the time, their energy efficiency is still under 60% in the operational mode (calculating from the lower hydrogen combustion heat) [3, 4]. So far, the world's main research directions in this field are the high-temperature electrolysis and the polymer electrolyte membrane (PEM) electrolyzers [3]. With the former, efficiency improvement is achieved by making the heat to activate the water splitting. However, a high-temperature system requires power to be supplied as heat, which is often not easy. At the same time, the PEM electrolyzers operate under normal conditions, with a relatively high efficiency – around 60%, which is still lower than that required for an energy storage scheme; furthermore, nowadays the PEM electrolysis systems are more expensive than the alkaline electrolyzers [4].

Typically, direct current (DC) power is used in electrolysis; nevertheless, pulse DC voltage also can be used [5–7]. Bockris *et al.* [6] observed a long current

tail immediately after applying a voltage pulse to an electrolytic cell. Shimizu *et al.* [5] applied inductive voltage pulses (200 ns) to feed the electrolytic cell and found that the efficiency of electrolysis does not change with the applied power. In our earlier work [7], we also used inductive voltage pulses to compare different metals as cathode materials and found that the concentration of dissolved hydrogen grows faster on the metals with higher overvoltage of hydrogen evolution and its lower solubility.

In this work, it is shown that applying inductive voltage pulses to the electrolytic cell it is possible to effectively reduce the applied potential by separation of the charging current from the charge transition (Faradic) current in a hydrogen evolution reaction.

2. EXPERIMENTAL

In our experiments a self-made electrolytic cell was used (Fig. 1). It consists of a poly-acetal body with two stainless-steel (SUS316L) electrodes. One electrode is fixed, while the other is moving and connected to a micro-screw. Special sealing holes are formed to avoid the contact of micro-screw holder material with electrolyte. Before experiments, the sealing holes were filled with insulating mastic. The electrochemical cell is shaped as a hollow with a diameter of 40 mm, while steel electrodes present 2 cm² squares. Before experiments the electrodes were mechanically polished and washed with acetone and deionised water. As electrolyte, the KOH solution with concentrations 0.1–0.5 M was used. During experiments the distance between electrodes was varied from 1 to 5 mm.

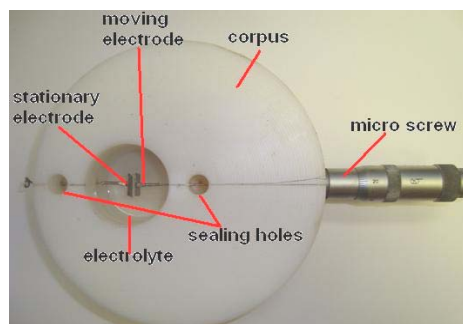


Fig. 1. Water electrolysis cell.

The inductive voltage pulses were generated in the electric circuit (Fig. 2) consisting of a pulse generator, a DC power source, a field transistor BUZ350, and a blocking diode [7, 8]. 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%). After obtaining inductive reverse voltage pulses, the primary winding of the transformer was powered with low-amplitude square voltage pulses. In the secondary winding (winding ratio 1:1) due to collapse of the magnetic field induced in the coil a very sharp high-amplitude inductive pulse arises that has the opposite polarity with respect to the applied voltage. The pulse of induced reverse voltage was passed through the blocking diode, and the resultant $\sim 1 \mu\text{s}$ wide high-voltage pulse was applied to the electrolytic cell. A two-

beam oscilloscope GWinstek GDS-2204 was employed to record the voltage (i.e. its drop on a reference resistance) and current in the circuit.

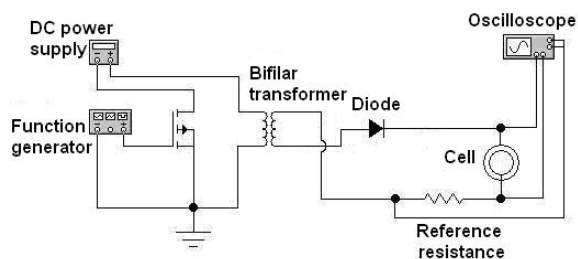


Fig. 2. Experimental circuit for generation of inductive reverse voltage pulses.

3. RESULTS AND DISCUSSION

After applying an inductive voltage pulse to the electrolysis cell with 3M KOH solution, a typical picture appears on the oscilloscope screen (Fig. 3). Oscillograms were taken at the electrodes spaced 3 mm apart. In all experiments the pulse repetition time of 50 μ s was chosen, and the direct pulse amplitude was set at 1 V. Both the voltage pulse and the current pulse had fast rising front. The voltage grew to a maximum, with the amplitude depending on the distance between electrodes, followed by a long discharge tail. As the tail end is reached, the current approaches zero, while the voltage stays around 2 V. A next pulse is identical to the previous one, and so on.

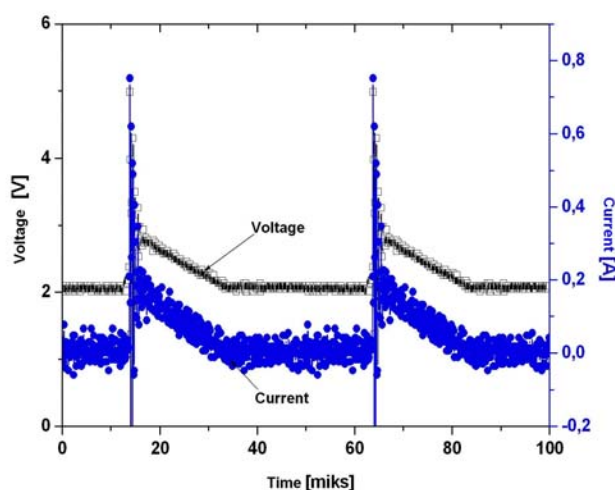


Fig. 3. Typical voltage and current pulses on the screen of oscilloscope when inductive reverse voltage pulses are applied to electrolytic cell.

The voltage and current oscillograms for 0.1, 0.3 and 0.5 M KOH solutions (Figs. 4–6, respectively) were measured at different distances between electrodes. As is seen in Fig. 4 (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 this distance is 1 mm. In the solution with higher concentration (0.3 M KOH, Fig. 5) the voltage pulse reaches only 3.5 V at the same distance between electrodes

(5 mm), and drops to 2.6 V when this distance is reduced to 1 mm. The more concentrated solution (0.5 M KOH, Fig. 6) gives pulses of lower voltage: 2.9 V at the distance of 5 mm between electrodes, and 2.4 V at this distance being 1 mm. The peak values of current do not change significantly in dependence on the electrode spacing and solution concentration, while the length of the discharge tail is changing, suggesting that the largest charge is transmitted in the most concentrated solution.

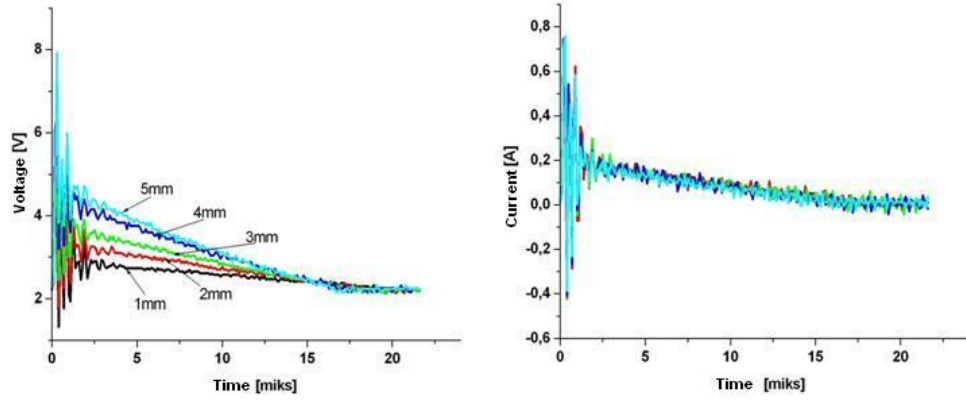


Fig. 4. Voltage and current oscillograms for a two-electrode cell in 0.1 M KOH solution.

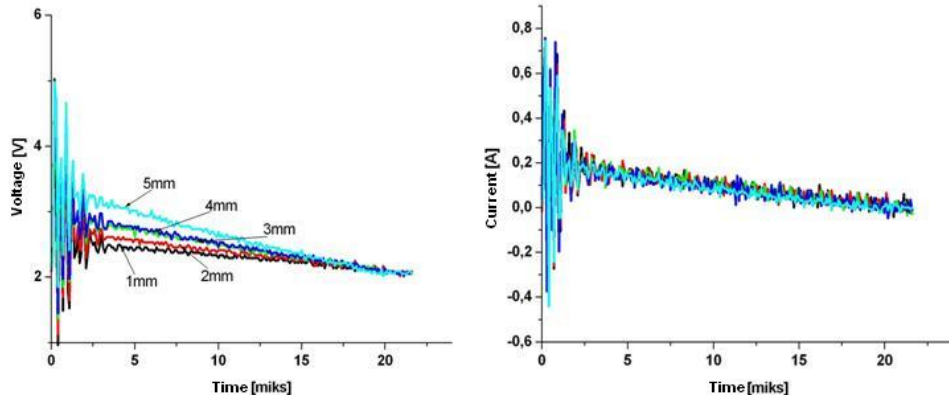


Fig. 5. Voltage and current oscillograms for a two-electrode cell in 0.3 M KOH solution.

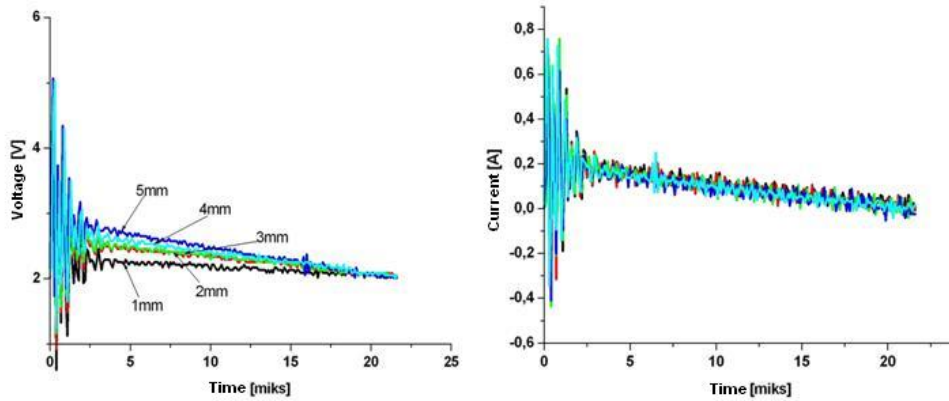


Fig. 6. Voltage and current oscillograms for a two-electrode cell in 0.5 M KOH solution.

The pulse generation scheme in Fig. 2 clearly shows that the nature of a high-voltage pulse created in the transformer is reactive. This is so since the amplitude of an active pulse in the circuit's primary side does not exceed 1 V, whereas in its secondary side the voltage exceeds 2 V and is reversed due to the blocking diode in the circuit. The reactive pulse amplitude depends on the quality factor (Q) of the capacitive element. A capacitor with a large leak is unable to keep high amplitude of the inductive voltage pulse, while from Figs. 3–6 it is seen that the amplitude of a reverse pulse is higher than that of a direct pulse.

A typical equivalent scheme of an electrochemical cell contains a parallel circuit with a capacitor and a resistor, which is series-connected with another similar parallel circuit. The former circuit reflects the geometrical capacity of the cell and the resistivity arising from oriented molecules, whereas the latter represents the capacity of a double layer on the electrode/electrolyte interface and the resistivity due to Faraday's hydrogen evolution reaction. Since the geometric capacity is much smaller as compared with the double-layer capacity, the first parallel circuit makes a little contribution to the overall picture.

When the voltage pulse is applied to an electrochemical cell, the double-layer capacity is charged, and this charging current is registered. In parallel, when the voltage on the double (electrode/solution) layer exceeds the red-ox normal potential, the conditions will be provided for the appearance of a Faradic current. The oscillograms in Figs. 3–6 evidence that the Faraday current has no time to arise within the first microsecond, and thus the amplitude of the reactive pulse is increasing. In other words, in the first microsecond the cell behaves like a capacitor with high Q . The more concentrated the solution is, the higher capacity of the double-layer capacitor; therefore, at the same charge the voltage will be reduced (cf. Figs. 4–6). As the distance between the electrodes decreases, the double-layer capacity is rising even faster, since in this case the voltage peak decreases significantly.

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

It has been inferred by Shimizu *et al.* [6] that when an active voltage pulse is applied to the water electrolysis cell the charging takes up too much of the pulse-time tracking. This does not favour the pulsed electrolysis as compared with the conventional DC electrolysis. However, if a reactive inductive voltage pulse is applied, the mentioned disadvantage is transformed into an advantage, because after such a short charge period the discharge tail appears to be several times longer than that of the pulse length taken to infinite resistance.

4. CONCLUSIONS

Application to water electrolysis cell of very short voltage pulses arising from disruption of current in the primary circuit gives rise to fast cell charging in the secondary inductive circuit which transforms into a relatively slower discharge tail. The authors believe that such kinetics can occur when the electrochemical

charging process in the cell is separated from a faradic charge transfer process. At applying a sharp pulse the water electrolysis cell behaves as a capacity with a high Q-factor, while the Faradic discharge manifests itself by a discharge tail, when the power stored during cell charging in short pulse is used.

The proposed power scheme for generation of inductive voltage pulses allows the power of the primary circuit to be reduced, thereby reducing the total power necessary for electrolysis. It is demonstrated that the electrolysis of water and water-alkali solutions can be achieved with the input voltage being as low as 1 V. A great advantage of the proposed pulse electrolysis is the fact that the charging of electrolytic cell lasts for a relatively shorter period than the following discharge using the previously stored energy.

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

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Kopsavilkums

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.

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