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Could one achieve a self-recharging double layer capacitor?

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The aim of this short communication is to initiate an open discussion topic, besides the main practical challenge, for electrochemists, electrical engineers or other scientists alike, to investigate and scrutinize whether or not it is possible to obtain, from theoretical point of view, as well as from the practical one, a self-recharging double layer capacitor. This topic has been suggested by the reconditioning in Romania of so-called Karpen's pile, exhibited in the Romanian National Technical Museum "Dimitrie Leonida" in Bucharest. This device provides power continuously (without any external source) to an electric motor, which was the main reason to be considered for a long period of time as a so called, allegedly, "*perpetuum mobile*". Recently, the mechanism of electrical energy generation in Karpen's pile has been elucidated in a paper published in Revista de Chimie. Moreover, this paper put forward a well-founded demonstration that Karpen's pile is in fact a self-recharging capacitor and not at all a *perpetuum mobile.*

Key words: self-recharging capacitor, Karpen's cells.

INTRODUCTION

The Romanian National Technical Museum "Dimitrie Leonida" in Bucharest has in possession and displays a device known as "*Karpen's Cell*", which sparked numerous controversies over the last 60 years as it gives the impression that it provides endless energy in the external circuit, apparently in the absence of the characteristic chemical processes or any energy transfer from the surrounding environment. The controversy is even greater in comparison with the *Oxford Electric Bell* or *Clarendon Dry Pile* in operation since 1840 [1,2], as for this type of pile the issue evolves around the fundamental questions "Does it contradict or not the 2nd Law of Thermodynamics? Is it or not a 2nd degree *Perpetuum mobile*?"

Nicolae Vasilescu-Karpen (1870-1964) was a professor at the Politehnica University of Bucharest and its Chancellor/Rector for more than 20 years (1920-1940). He approached the research of this type of cells that bears his name since the second decade of the twentieth century, patenting the first models in 1924 [3].

From the very beginning, *Karpen's cells* were shrouded in an aura of mystery even by their author, who said that their operation contradicts the 2nd principle of thermodynamics [4], that they are a form of a 2nd kind *perpetuum mobile*, fact also advocated until recently by other authors [5].

Despite numerous practical demonstrations made by Karpen with the *Karpen's cells* (Paris, 1922) [6, 7] or theoretical lectures in Bucharest followed by publications [8], the subject had a small audience and received little support in the international scientific community because these devices were associated with the idea of a perpetuum mobile. Strictly speaking, *Karpen's cells* are unpartitioned electrochemical systems, tightly sealed, containing two inert metals (gold, platinum) or graphite, immersed into the same electrolyte solution (sulfuric acid solution, pure sulfuric acid) or even in distilled water, containing small amounts of dissolved oxygen (Oxygen *Karpen's Cell* - KOx), or hydrogen (Hydrogen Karpen's Cell - KHy). One maintains in both cases, above the solution surface, a layer of air or hydrogen, at a low partial pressure, about 100 Pa [9].

In a number of public lectures (held in Timisoara and Bucharest, 2016) and in two recently published papers [10, 11], we demonstrated that *Karpen's cells* are in fact electrochemical double layer capacitors, which after a discharging process recharge themselves due to a series of endothermic spontaneous processes taking place in the above said systems, leading to a cooling of the electrolyte

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solution. The occurrence of a temperature gradient between the electrolyte solution and the surrounding external environment determines the generation of a heat flow rate bringing the solution temperature close to the ambient temperature, compensating for the heat loss induced by the spontaneous recharging process. As a result, *Karpen's cells* convert heat supplied from the surrounding environment into electrical energy powering an external resistive circuit made of a micromotor suspended by a torsion wire, thus *Karpen's cells* acting as a heat to electricity (pseudo)converter.

The most significant results achieved by Nicolae Vasilescu-Karpen were in the case of KOx cell, made of a platinized gold electrode (AuPt) of 2000x6x0.4 mm, coiled onto a glass substrate (positive terminal), placed symmetrically between two shiny gold electrodes of an identical geometry, and pure sulfuric acid as electrolyte. KOx cell powers intermittently a micromotor: the electric motor is connected with the cell for a period of time of 0.5 s, followed by a break of 18 s (Fig. 1) [12].

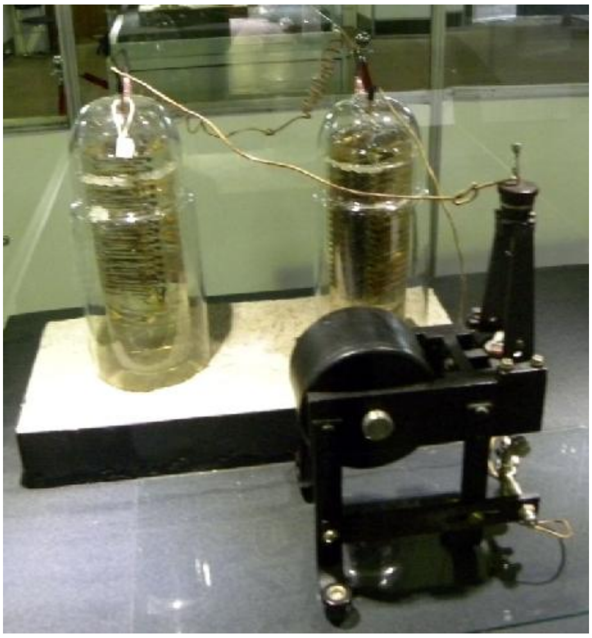

Fig. 1. KOx Karpen's cell, (Courtesy of Romanian National Technical Museum *Dimitrie Leonida,* Bucharest).

The maximum current intensity measured during the contact phase is 17.5 µA, the terminal voltage decreasing to 0.02÷0.03 V. After 18 seconds recovering period, the terminal voltage reaches approximately 0.25 V [12].

THE MECHANISM OF ELECTRICAL ENERGY GENERATION IN KARPEN'S CELLS

Being given the favourable conditions for KOx cell, namely the low partial pressure of oxygen above the electrolyte solution, the corresponding amount of oxygen dissolved in sulfuric acid solution is very small. Consequently, the AuPt and Au electrodes surface will not be saturated with molecular or atomic oxygen (resulting from the dissociation of O_2 molecules). We may admit that in this situation, the fractional occupancy of the adsorption sites of electrodes with oxygen is proportional to the strength of metal–oxygen bond. Since Pt–O bond is stronger than the Au–O bond [13], the activity of the absorbed oxygen on AuPt electrode is higher than the activity of oxygen adsorbed on Au. If the electrolyte solution and the electrode material are of an advanced purity degree, one shall establish an equilibrium at the interface, given by the reversible reaction:

$$O_2 + 4H_3O^+ + 4e^- \rightleftarrows 6H_2O \qquad (1)$$

Considering the values of the oxygen activity specifically at the two electrode surfaces, the reversible process equilibrium (1) on AuPt is tilted more towards the right side comparing to that taking place on Au, in accordance with the Châtelier-Braun principle. Therefore, in order to achieve a steady state equilibrium, the AuPt electrode will yield more electrons than the Au electrode, i.e. AuPt will become the positive terminal and Au - the negative terminal of this electrochemical system, so we may write the following:

$$(+)AuPt(O_{2(ads)})/H_2SO_{4(aq)},\ O_{2(aq)}/Au(O_{2(ads)})\ (-) \qquad (2)$$

It is well known that the charging process of the above said two metal/solution interfaces is a spontaneous non-faradic process that leads to the formation of an electric double layer [14]. When the electric micromotor is connected to the two cell terminals, a current is passing through the external circuit that determines the micromotor to move in accordance with the above described mechanism. In a matter of fractions of a second (0.5 s), the electrochemical system will discharge its load, the electrochemical potentials of the two electrodes becoming approximately equal. This behaviour allows us to affirm that the considered electrochemical system is, at limit, **a capacitor** and **not a galvanic cell,** as it was previously believed. During the discharge phase, the electrons pass from the Au electrode through the external resistive motor circuit, reaching finally the AuPt electrode.

When the external circuit is switched off, the AuPt electrode will have an excess of negative electric charges (electrons), while the Au electrode will have an excess of positive electric charges (electrons deficit). This actually means that, in order to restore the chemical equilibrium balance between the two metal - electrolyte solution interfaces, reaction (1) will take place on the AuPt electrode, the equilibrium being shifted towards favouring the reduction process (3):

$$O_{2(ads\ AuPt)} + 4H_3O^+ + 4e^- \rightarrow 6H_2O \qquad (3)$$

At the same time, the conjugated oxidation process takes place on the Au electrode:

$$6H_2O \rightarrow O_{2(ads\ Au)} + 4H_3O^+ + 4e^- \qquad (4)$$

The overall reaction taking place during the recharge phase of the electrochemical system (2) is obtained by adding up reactions (3) and (4):

$$O_{2(ads\ AuPt)} \rightarrow O_{2(ads\ Au)} \qquad (5)$$

It means that the capacitor charging consists formally by passing the molecular oxygen from AuPt electrode to the Au electrode. As Pt–O bond is stronger than that of Au–O, reaction (5) will be an endothermic one, therefore the capacitor receives during the charging process external thermic energy, which is transformed into electrical energy during the discharge phase. The heat flow rate directed from the outside towards the capacitor has its origin in the very small temperature gradient, taking into account the extremely low electrical current generated during the discharge phase. The electrical energy generated is then converted using the external circuit into a mechanical work using the micromotor and heat, given the passage of a current through the resistive element, known as Joule effect, the energy losses being given not only by the friction between the moving mechanical parts, but also by the frictions with the air; some of the energy is also lost by the heating of the external resistive element by Joule effect, as explained above. Consequently, the electrochemical system is considered a self-charging capacitor whose operation does not contradict the 2nd thermodynamics principle, so it is not a 2nd kind *perpetuum mobile.* Since the charge is done by non/faradic processes, KOx cell is a double layer electric capacitor and not a galvanic cell. Similar processes take place in hydrogen *Karpen's cell* (KHy). Preliminary attempts to achieve a *Karpen's cell* with hydrogen at normal pressure, close to the atmospheric pressure, were doomed to failure because the electromotive force obtained was close to zero.

However, changing the conditions leads to a considerable voltage, namely when the cell is fitted with platinum and shiny gold electrodes having the same size (20X30 mm). The basic solution was diluted sulfuric acid (0.30%), and the partial pressure of the hydrogen above the solution was 918 Pa, which means that the dissolved hydrogen concentration in the solution is very low. At temperature of 20°C, the cell terminal voltage of KHy was 0.15 V [9]. Under these conditions, the metals surface will not be saturated with hydrogen and the activity of the adsorbed hydrogen on the metals surface will be proportional to the energy of metal–H bond. According to the available data, Pt–H bond energy is higher than the Au–H bond energy [15].

For simplification, let us assume that hydrogen adsorbed on the metals surface, taking into account its reduced concentration, is found in its atomic state [16, 17]. The reversible equilibrium established is given by equation 6:

$$H + H_2O \rightleftarrows H_3O^+ + e^- \qquad (6)$$

It is obvious that on Pt the equilibrium (6) will be more shifted to the right than on Au. As a result Pt will acquire an excess of electrons, while Au will have a deficit of electrons, which means that Pt will be the negative terminal of KHy cell, and Au, the positive one. Short-cutting the electrodes leads to a flow of the excess electrons from Pt towards Au, performing a mechanical work in the external circuit, while the electrodes terminal voltage will drop quickly. When the external circuit is open, at the Pt electrode will occur reaction (6) shifted towards the oxidation:

$$H_{(ads\ Pt)} + H_2O \rightarrow H_3O^+ + e^- \qquad (7)$$

and the conjugated one, the reduction reaction, will take place on the Au electrode:

$$H_3O^+ + e^- \rightarrow H_{(ads\ Au)} \qquad (8)$$

The overall reaction is given by the transport of adsorbed hydrogen from Pt to Au electrode:

$$H_{(ads\ Pt)} \rightarrow H_{(ads\ Au)} \qquad (9)$$

As Pt–H bond is stronger than Au–H bond, reaction (9) is an endothermic one, therefore, similarly to the case of KOx cell, KHy cell extracts energy during the recharging process from the surrounding environment.

RESULTS AND DISCUSSION

Development prospects for Karpen's Cells

KOx and KHy cells are difficult to study because of the practical problems associated with their manufacture: tightly sealed installation, low partial pressure of O_2 and H_2 and difficult to control. This is precisely why we bring forward the proposal of another type of self-charging double-layer capacitor, made of two different electrodes immersed in a solution of potassium iodide, containing also small amounts of iodine-iodide. The metals used as electrodes have to meet the following condition: the difference between metal–I energy must be as larger as practically possible. If the electrolyte solution is of a high purity and does not contain dissolved gases, at the interface metal-electrolyte solution the following equilibrium will be established:

$$I_3^- + 2e^- \rightleftarrows 3I^- \qquad (10)$$

As the equilibrium constant for the reversible reaction $I_2 + I^- \rightleftarrows I_3^-$ is high (698 at 25°C [18]), we may consider that I_2 is found in the solution only as I_3^-.

If the concentration of I_3^- ions is reduced and that of I^- ion is in large excess, then the degree of adsorption on the electrodes surface will depend on the strength of metal$-I_3^-$ bond, meaning that the I_3^- ions activity at the interface will also be depending on the strength of metal$-I_3^-$ bond. Considering now that the metal$-I_3^-$ bond is noticeably stronger for M_1 than for M_2, let us examine now the Galvani potential profile alongside the electrochemical system:

$$M_1(I^-_{3\,(ads)})/\ I_3^-,\ I^-,\ K^+(aq)/M_2(I^-_{3\,(ads)}) \qquad (11)$$

Depending on the I_3^- ions concentration, one may have the following cases, presented in Fig. 2:

A - I_3^- ions concentration is high enough, so that the surfaces of both electrodes are to be saturated with I_3^- ions, namely the ratio of I_3^- and I^- concentrations at the interface is to be equal to the ratio of their concentrations from the electrolyte solution. In this case, the absolute potentials of the two electrodes are equal to each other, and the terminal voltage of the considered electrochemical system is zero (Fig. 2, profile A);

B – I_3^- ions concentration is high enough, so that the surface of metal M_1 is to be saturated with I_3^- ions, but, at the same time, the concentration of I_3^- ions is low enough so that on the surface of metal M_2 there is a deficit of I_3^-ions. In this situation, the absolute electrode potential of M_1/S (S - the electrolyte solution used in the system) is unchanged comparing to the case of A, but the absolute potential electrode M_2/S will be shifted towards more negative values (Fig. 2, profile B);

C - the concentration is so small that one may not reach the saturation level on both electrode surfaces, which means that the potentials of both electrodes will be shifted towards more negative values, however, of a smaller magnitude in the case of M_1/S and bigger in the case of M_2/S (Fig. 2, profile C).

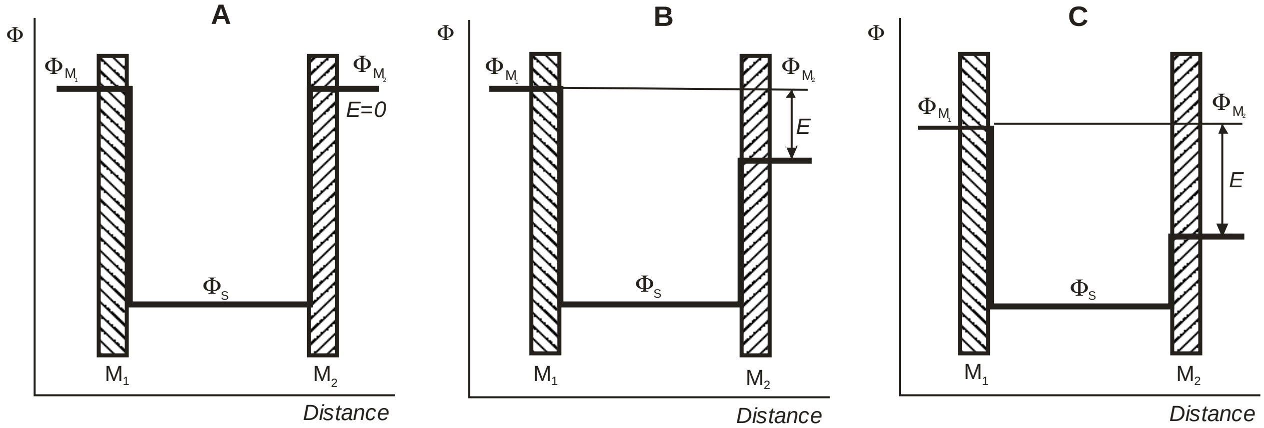

Fig. 2. The profile of the internal Galvani potential alongside the electrochemical system (11).

As one may easily see from Fig. 2, in cases B and C a voltage drop is produced between metals M_1 and M_2 due to the different charge of the electric double layer at the interface M_1/S and M_2/S. This is one of the reasons that the above discussed electrochemical system (11) may be considered as being a capacitor, where M_1 is the positive terminal and M_2 is the negative terminal. The absolute potentials may now be expressed using the Nernst relationship:

$$\Delta\Phi_{M_1/S} = \Delta\Phi^0_{I_3^-/I^-} + \frac{RT}{2F}\ln\frac{a_{I_3^-(ads\,M_1)}}{a_{I^-(S)}} \qquad (12)$$

$$\Delta\Phi_{M_2/S} = \Delta\Phi^0_{I_3^-/I^-} + \frac{RT}{2F}\ln\frac{a_{I_3^-(ads\,M_2)}}{a_{I^-(S)}} \qquad (13)$$

As a result, the electromotive force between the two electrodes is given by:

$$E = \Delta\Phi_{M_2/S} - \Delta\Phi_{M_2/S} = \frac{RT}{2F}\ln\frac{a_{I_3^-(ads\,M_1)}}{a_{I_3^-(ads\,M_2)}} \qquad (14)$$

From relation (14) one may notice that E is dependent on the ratio between the energies of the metal–I_3^- bonds; the higher this ratios, the greater the E.

The equivalent capacitance C for the electrochemical system (11) shall be given by:

$$\frac{1}{C} = \frac{1}{C_1} + \frac{1}{C_2} \qquad (15)$$

and hence:

$$C = \frac{C_{M_1/S} \cdot C_{M_2/S}}{C_{M_1/S} + C_{M_2/S}} \qquad (16)$$

where $C_{M_1/S}$ și $C_{M_2/S}$ are the individual capacitances of the two metal/solution interfaces. By inserting a micromotor connected to the two electrodes, in order to produce a mechanical work in the external circuit, the electrons shall pass from M_2 to M_1, so that, shortly, the electrodes potentials shall become equal, namely the capacitor will be discharged.

Opening the external micromotor circuit, the equilibrium of reaction (10) will be shifted towards the reduction side, due to an excess of electrons:

$$I^-_{3\,(ads\ M1)} + 2e^- \rightarrow 3I^-(aq) \qquad (17)$$

At the same time, on metal M_2 the reaction will take place shifted towards the oxidation side:

$$3I^-(aq) \rightarrow I^-_{3\,(ads\ M2)} + 2e^- \qquad (18)$$

Equation (19) represents the overall reaction describing the process taking place when the capacitor is charged and it is obtained by adding up reactions (17) and (18):

$$I^-_{3\,(ads\ M1)} \rightarrow I^-_{3\,(ads\ M2)} \qquad (19)$$

The charging process of the capacitor depicted in (11) is similar with the above discussed systems KOx and KHy, being an endothermic one. As a result the electrochemical system (11) transforms the thermal energy taken from the surrounding environment during the charging phase into a mechanical work and heat using the external circuit during the discharge phase. It is obvious and easy to understand that the electrochemical system (11) will operate intermittently, as the capacitors $C_{M1/S}$ and $C_{M2/S}$ charging takes place with a limited rate. However, such a capacitor is useful to provide intermittent power/signal in isolated environment, such as the airplane "black box", in electronics for intermittent component excitations (e.g. transistors), in communications systems for blocking the DC signal on the transmission line, or as independent, intermittent signal generator with potential applications in securing a certain perimeter.

A particularly interesting feature of such devices with potential application in evaluating their state of charge/health is linked to the fact that one needs here only straight simple measurements to do this; as the total energy contained in these devices depends only on their capacitance (in fact a constant value) and on the voltage values, one may measure these variables easily and hence the simplicity in determining the total charge or energy stored or left in these devices. With nowadays state of the art electronic gauges, this assessment is easy to be implemented and carried out.

The above presented capacitors operate at room temperature, being limited only by the liquid state of the electrolyte solutions. One may also find suitable similar capacitors of this type, able to operate at low or high temperatures, depending on the nature of electrolytes and solvents used such as: molten salts, ionic liquids, organic solvents.

A novel proposal for a self-charging double-layer capacitor consists of two different metals M_1 and M_2, immersed in a molten mixture of KI and LiI, containing also a small amount of I_2. The melting temperature of the eutectic system LiI-KI (LiI 63.2%, 36.8% KI) is 287°C, and that of the individual molten salts is 469°C (LiI) and 681°C (KI) [19]. As a result, such a capacitor is operational in a wide range of temperatures. Triaca *et al.* have shown that in molten salts it is more likely to have the participation of I_2 species in establishing the electrochemical equilibrium in the presence of I^- ions, rather than that of I_3^- [20].

Such a capacitor is best represented by the following electrochemical chain:

$$M_1(I_{2(ads\ M1)})/\ I_2(l),\ LiI(l),\ KI(l)/M_2(I_{2(ads\ M2)}) \qquad (20)$$

Metals M_1 and M_2 are selected so that there is a large difference in the energy of metal–iodine interactions for the two metals. As there are no available literature data regarding the metal-iodine energies in molten salts, the selection of M_1 and M_2 is carried out so that there is a significant difference between the electron work function of the two metals. Examples of metal with an appreciable electron work function are: Pt (5.65 eV), Ir(5.27 eV), Ni(5.15 eV), Pd(5.12 eV), Au(5.1 eV), Co(5.0 eV), which may be associated in a self-charging dual-layer capacitor with metals with low electron work function such as Ta(4.25 eV), Ag(4.26 eV), Nb(4.3 eV), Ti(4.33 eV), Fe(4.5 eV) [21].

Let us consider that the electron work function for the metal M_1 is smaller than that for the metal M_2. The equilibrium established at the metal-molten salt interface will be given by the equation:

$$I_2 + 2e^- \rightleftarrows 2I^- \qquad (21)$$

If we have a low concentration of I_2 in the molten salt, the equilibrium established on metal M_1 will be given by (21) and it will be shifted more towards the right side than the similar equilibrium established on metal M_2, resulting that, for the electrochemical system (20), M_1 will be the positive terminal and M_2 - the negative terminal.

The charge and discharge processes taking place in the capacitor (20) are similar to those occurring in the capacitor (11) made in an aqueous solution.

One of the main characteristics of the above presented mechanism is that the process of storing the electrical charge does not involve alterations of any chemical bonds of the electrode/electrolyte and hence a life-time expressed in charge-discharge cycles limited only by the materials properties. This type of devices may be used for just a fraction of the stored charge or to the full extent, frequently or infrequently, without worrying about the so called „memory effect", being particularly useful (with adequate capacity considered) for back-up power sources such as UPS (uninterruptible power supplies) systems or in vehicles powered by hybrid systems.

Moreover, as they have an increased tolerance to low, as well as high temperatures, one may envisage potential applications (such as space explorations, volcanic research, deep see investigations, high or low temperature industrial processes) where such temperatures are unavoidable and unchangeable, and the classical galvanic cells will irreversibly break down in no time (the decrease of the electromotive force with temperature, electrolyte boiling or freezing etc.).

CONCLUSIONS

The foregoing demonstrations entitle us to state unambiguously that Karpen's, such as KOx and KHy can be considered as self-charging double-layer capacitors, such devices being therefore perfectly achievable from both theoretical and practical point of view. The affirmative answer to the question set in the title raises however some additional issues: what is the capacitance and the efficiency of such electrical power sources?

One of the main advantages of the capacitors in comparison with the classical galvanic cells is the high stability of the electrical charge stored, as in the case of capacitors we no longer talk about a chemical reaction and hence to the absence not only of the primary reactions, but also of the secondary unwanted reactions which lead to a decreased current efficiency. The drawback here is the reduced time due to the self-discharge phenomenon, so one should pay particular attention when considering such systems for long term energy storage.

Devices as the above presented ones, in existence and operating for more than 50 years, should be looked upon not as a simple scientific curiosity, but as an inspiration source for open-ended novel potential applications.

The necessity for advanced systematic studies in this particular field of such interesting revisited devices calls for interdisciplinary research teams (physicists, electrochemists, electrical and materials engineers), as this issue tends to be more than just a simple theoretical curiosity, overcoming long time ago the theoretical stage.

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ВЪЗМОЖНО ЛИ Е ДА СЕ ПОСТИГНЕ САМОСТОЯТЕЛНО ПРЕЗАРЕЖДАНЕ НА ЕДИН ДВУСЛОЕН КОНДЕНЗАТОР?

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(Резюме)

Целта на това кратко комюнике е да инициира отворена дискусия по темата наред с основното й практическо предизвикателство за електрохимици, електроинженери и други учени, за да се изследва и проучи дали е възможно както от теоретична, така и от практическа гледна точка използването на самостоятелно презареждащ се двуслоен кондензатор. Тази тема е предложена в Румъния за преустройство на така наречената „батерия на Карпен“, изложена в Румънския националния технически музей "Димитрие Леонида" в Букурещ. Това устройство осигурява непрекъснато захранване (без външен източник) на електрически мотор, което е основната причина да бъде разглеждано като така нареченото "перпетуум мобиле". Наскоро механизмът на производството на електрическа енергия в батерията на Карпен бе изяснен в статия, публикувана в “Revista de Chimie“. Тя предлага и една добре обоснована демонстрация на батерията на Карпен като самостоятелно презареждащ се кондензатор, а не като „перпетуум мобиле“.