Applications of Electrochemical Elements in Systems of Artificial Intelligence

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Abstract: Contemporary systems of artificial intelligence transduce information primarily by application of electronic, rather than nonelectronic, devices. Recently, numerous reports have been published about the research and development of electrochemical transducers, which are fully compatible with the technology of solid-state electronics and can provide substantial benefits in the circuit design. In nature, intellectual functions are carried out on the basis of electrochemical and chemical mechanisms of information transfer, such as occurs in the human brain. Artificial intelligence systems allow one to replicate and to amplify the various functions inherent in nature. This article describes the main results and the probable prospects of technologies which may permit one to carry out certain intellectual functions by means of technical solutions based on the application of electrochemical transducers of information.

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1. Introduction

Systems of artificial intelligence are the main goal in the development of modern computer technologies. Their construction requires application of different transducers, most of which are based on electronic mechanisms. In addition to electronic elements, there are also optical, thermal, electro-magnetic, electromechanical, and other elements that are based on physical mechanisms. However, there is a much less well known transduction element which employs an electrochemical mechanism using electrolyte-containing cells. The application of such elements in modern devices is slowly but steadily increasing. Recently, there also have been numerous reports on the research and development of electrochemical cells based on solid electrolytes, which are fully compatible with the technology of modern solid-state electronics and can provide substantial benefits in circuit design.

In nature, the intellectual functions of various organs are carried out on the basis of electrochemical and chemical mechanisms. The prime example of one such organ is the human brain. Artificial intelligence systems can potentially replicate and amplify various functions inherent in the brain. There is a common assertion that the brain is an electrochemical computer. For example, Bill Seaman and Otto E. Rossler [1] formulated three possible approaches to the creation of an electro-chemical computer that performs the functions of the brain and submitted an abstract scheme of the arrangement of different modules in such a computer. Also, a mathematical consideration of the simplified electrochemical model of a brain was presented in the publications of T. Triffet and H. S. Green [2]. Therefore, the question arises to what extent the level of current research and development in the field of electrochemistry allows us to come nearer to creation of more advanced computer models capable of performing brain functions.

This article describes the main results and the probable prospects of technologies which may permit one to carry out certain intellectual functions by means of technical solutions based on the application of electrochemical transducers of information.

2. Electrochemical Memory Elements in Artificial Neural Networks

The first time the idea of creating systems that use electrochemical elements of memory appeared, it was in connection with the task of modeling neural networks. The concept of an artificial neural network was formulated by McCulloch and Pitts in 1943 in the form of mathematical and electrical models of biological neural networks. On the basis of such networks, development of adaptive computer systems began, and the most attractive of these systems was the perceptron.

Learning ability is usually achieved by storing data changes in neural networks. For this purpose, it is necessary to apply elements in electrical circuits that have several requirements: a large number of storage levels, a reversible mechanism of their formation, a long memory retention time, and no consumption of energy during storage. To fulfill these requirements, Bernard Widrow in 1960 applied a new electrochemical element, which he called a memistor.

The memistor, designed by B. Widrow and T. Hoff [3], was a sealed cell made of polymeric material in which the resistance of the graphite electrode reversibly varied in the range of 1 to 100 Ohms by electrochemical deposition or dissolution of copper. The size of resistance was achieved as a result of passing of multidirectional impulses in an operating circuit. Resistance was measured in a control circuit. The cell had three electrode terminals, one of which was common to both circuits. As described by the authors, their memistor allowed to achieve up to 1000 storage levels.

In the mid 1960's, due to the popularity of works on the creation of perceptrons, advanced models of memistors were developed. They differed from Widrow's memistor in that electrodeposition of copper was carried out on a cylindrical micro-electrode, which was a platinum wire 5 microns in diameter [4]. These microelectrodes were made on the basis of Vollaston platinum filaments. Theoretical calculations and experimental studies have shown that due to the cylindrical configuration of the diffusion layer such microelectrodes can increase the speed of electrodeposition and dissolution of copper by 10 times [5]. Additionally, it also allowed a linear dependence between measured resistance and quantity of the electricity forming storage levels. The mentioned works were carried out in a number of laboratories with coordination by the Institute of Electrochemistry of the Academy of Sciences where the scientific management was carried out by P. D. Lukovtsev [6].

In the 1960's, increased attention to electro-chemical memistors was given due to the fact that they allowed the construction of the most efficient artificial neural networks for perceptrons, in comparison with other circuit components. However, later publications showed the fundamental limitations of artificial intelligence based on percep-trons. Furthermore, it was discovered that memistors in which the resistive layer is formed by means of electrodeposition and electrodissolution of copper aren't completely reversible, since in the course of cycling the electrode accumulates a quantity of copper which does not participate in creation of the resistive layer [7]. As a result, work on memistors based on solutions of copper lost relevance. After that, in accordance with the general concept of solid-state microelectronics, the development of memistors was carried out on the basis of solid electrolytes.

The first sufficiently detailed report about creation of such a component appeared in 1990. The authors called this the solid-state thin-film memistor [8]. According to the authors' description, the adjustable resistive layer was formed by tungsten acid, which was obtained from tungsten oxide. The necessary hydrogen ions were delivered by means of reversible electrochemical transfer from a layer consisting of a hydrated oxide of hexavalent chromium. This process allowed the memistor to change resistance from 10^5 to 10^9 Ohms and provided a stable memistor lasting several months. The authors also point out the need for further research of their electrochemical system to avoid gassing at the electrodes. From the description of the experiment provided in their article, it follows that gas emission could arise owing to existence of ions of chromic and tungsten acids: i.e. the chosen system, despite the name, was not solid enough.

In 2005, V. Yerokhin,T. Berzina and M.P. Fontana published an article about the development of an electrochemical device on the basis of thin layers of polyaniline and polyethylene oxide [9]. The insulating substrate was coated with a 5-10 micron layer of solid electrolyte composed of polyethylene oxide and lithium chloride. Finally, above the coated layer, a very thin layer (~48 nm) of polyaniline was deposited. The component had three electrode terminals. Electric resistance of a polyaniline layer changed as a result of the transfer of lithium ions into this layer and the subsequent electrochemical redox reaction. According to the authors' assertion, the device showed stable and well reproducible electrical characteristics. The article points to the possibility of miniaturization of the device through the use of electron-beam processing for the purpose of creating a nanoscale configuration. In addition, it was noted that the resulting dependence of the conductivity on the exposure time of voltage can be the basis for the realization of adaptive polymer structures.

At present, most attention is given to the research and development of two-electrode electro-chemical elements functioning on the basis of reversible redox reactions and ion transport in the nanolayers of insulators and semiconductors. Several works have reported the studies of reversible changes in electrical resistance when thin (20 - 300 nm) layers of oxides of aluminum, silicon, titanium, and other fine-crystalline and amorphous insulators were exposed to an alternating voltage. These multiple works appeared in the 1960's, i.e., almost simul-taneously with the first information about memistors. They reported research of ion transition in various "metal-insulator-metal" systems. Changing of elec-trical resistance of an insulator resulted from injection or extraction of ions from adjacent metals.

In 1971, Leon Chua published an article which, proceeding from the principle of symmetry, theoretically showed a device which expresses dependence between charge and magnetic flux [10].

Along with the resistor, the capacitor and the inductor, he posited a "fourth fundamental circuit element" for complete characterization of circuit properties. He called it "memristor" as a result of reduction of the words "memory + resistor". In subsequent articles, he also introduced the concept of "memristance," defined as the property of an electronic element to remember its last resistance before being switched off. During the publication of his theoretical conclusions, L. Chua did not know about prior research and the memistor proposed by B. Widrow with a very similar name.

Memistor and memristor are similar elements, but not equivalent in their circuit design features [11, 28]. In the same article, L. Chua mentioned that creation of a memristor will allow the realization a number of unique properties which can't be reached in circuits consisting only of resistors, capacitors and inductors. However, in spite of the theoretical appeal of the memristor, the subsequent practical development of this element faced serious technological problems, which for a long time didn't even allow the creation of prototypes suitable for use in computer structures.

In the components considered earlier, memory process was carried out on the basis of an electrochemical mechanism. In memristors which are developed now, the process of memorizing is carried out on the basis of various mechanisms. According to the classification of R. Vaser [12], the process of memorizing can be performed using the following effects: ferroelectric, electrostatic, phase transitive, thermochemical, magneto-resistive, nanomechanical, by intramolecular reorganization, by changes in valence, or by electrochemical metallization. Thus, only the two last effects can be considered within the electrochemical mechanism. In particular, these two have attracted the largest amount of attention by researchers and developers as evidenced by the latest publications.

In 2008, D. B. Strukov, G. S. Snider, D. R. Stewart and S. Williams from authoritative computer firm Hewlett-Packard, reported about the development of a model of memristor on the basis of the Pt–TiO_2/TiO_{2-X}–Pt system [13]. They also suggested a more accurate mathematical model of the developed memristor, as compared to previously presented works of L. Chua. This model was in good consistence with their experimental results. In establishing their model, the authors proceeded from the fact that the memristance effect occurs in solid-state, nanoscale systems in which electronic and ionic transport is coupled under "external bias voltage." They note that the application of the external bias voltage produces a hysteretic behavior in the change of the current. According to the authors' proposal, this hysteresis requires atomic rearrangement that modulates the electronic current. The total memristor's resistance is defined by two components: a semiconductor layer with high dopant concentration of positive ions which has a low resistance (R_{on}) and the remainder layer with negligible ion concentration of the same ions and much higher resistance (R_{off}). Application of an external voltage causes the displacement of the boundary between the layers. On the basis of the accepted mechanism, the authors derived an equation defining the dependence of memristance from the total thickness of both layers, average mobility of dopants, and charge as a function of time. According to the derived equation the memristance becomes larger in absolute value for higher dopant motilities and for smaller film thicknesses of the semiconductor. Memristance becomes more important for understanding the electronic characteristics of any device as the critical dimensions shrink to the nanometric scale. Another team of developers of the same memristor explained that the role of dopants was carried out by positively charged oxygen vacancies [14]. In the subsequent publications about this development [15], one promising quality of the considered memristor is an exponential decrease of the energy spent during transition from high to low resistance, as current increases.

There are also other reports of memristor research using the transition metal oxides (VO, Ta_2O_5, Nb_2O_5, $SrTiO_3$) as a nanoscale layer and still other memristors based on the mechanism of charge transport, including migration, diffusion and ion valence change [16]. At the same time, there are a significant number of research and development proposals related to the "metal-insulator-metal" systems in which the mechanism of storing is caused by formation of subtle metallic filaments during electrochemical discharge of cations and their subsequent dissolution during the change of polarity [17]. Instead of insulators (such as SiO_2 or Al_2O_3) as the intermediate layer, some solid electrolytes (Ag_2S, Ag with dopants of $GeSe_2$) can be used. The cell of these memristors has two electrodes which are made from metals with different electrochemical properties. The first one, the active electode (AE), is made from electrochemically active metal with high conductivity Ag (for research) purposes or Cu (for broad consumption). The second electrode is an electrochemically inert counter electrode (CE) which is made from Pt (for research) or W (for broad consumption). Such memristors need a preliminary formative operation, during which porous channels are formed in the insulator. The existence of a predetermined pore structure provides further reproducibility of the formation and dissolution of metallic filaments. Speed of these processes depends on what type of material (an insulator or solid electrolyte) is used for creation of the intermediate layer. In certain cases, ion mobility can make a more essential contribution to memristance than the rate of discharge at the electrodes.

Various authors report about stability of constructed samples and compliance of sample parameters to essential operating requirements. For example, achieved switching speed corresponded to the nanosecond range. Meanwhile, in devices of memory applied now, these speeds are two orders lower. The projected time of memory retention, calculated on the basis of imitating tests at temperatures of 70 - 130^{0}C is ten years.

Modern computers are created on the basis of the materials whose properties are described by solid-state physics. These materials possess stability of electric properties in a rather wide range of temperatures and possess good reproducibility of functional parameters. Unlike them, information systems in nature are constructed on the basis of liquid electrolytes. Electrical properties of liquid elements are more significantly temperature dependent than that of solids. In addition, the changes in their electrical parameters, which happen frequently in the course of functioning liquid systems, have significantly poorer reproducibility than solids. Despite this, many researchers believe that the use of chemical and biological media, based on aqueous solutions and other liquids, is promising, as they allow one to create information systems with new functionality (such as for example the molecular and biological computers). In this regard, publications about research and development of new electrochemical transducers based on aqueous solutions continue to appear.

In 2000, V. N. Ur'ev, B. M. Grafov, A. V. Dribinskii and V. P. Lukovtsev published an article in which they demonstrated the realization of a multi-bit cell with a liquid electrolyte, where data are written by a successive cathodic deposition of alternating metallic layers with different electrochemical properties [18]. The written data was read in two ways: on the basis of successive controlled anodic dissolution of the multilayer structures or by recording the potential difference between the electrodes. Record/reading speed on 20 µm diameter electrode in a copper sulfate electrolyte was 10 Kbps. By reducing the diameter of the electrode, this speed can be increased by several orders of magnitude.

A more long term outlook to create molecular memory elements based on the application of liquid electrolytes has been reported as well [19].

3. Electrochemical Transistors Based on Organic Polymers in Artificial Intelligence Systems

Transistors are one of the key elements of electronic devices. Modern industry offers a large selection of different types of transistors, most of which are made on the basis of solid-state technology. Electrochemical transistors can be made of inorganic or organic materials. In patent literature, the variety of electrochemical transistors based on inorganic materials is classified as H01G9/26. However, the electrochemical transistors developed so far on the basis of inorganic semiconductors, have no essential functional or technological advantages and therefore can't compete with the transistors produced by widespread methods of solid-state electronics.

Industrial samples and technologies of electrochemical transistors creating new functionality for systems of artificial intelligence appeared as a result of creation of organic semiconductor materials and the development of methods of their application to nanoscale products.

Targeted research to develop polymeric semiconductors began in the 1950's. The most important stage of these studies was the development of halide polyacetylene in 1977, which has electric conductivity two orders of magnitude higher than previously known organic polymers [20]. Currently, polymers are created in a wide range of conductivity, from values corresponding to semiconductors (10^0 - 10^{-2} S/cm) to values approaching characteristics of metals (10^4 - 10^5 S/cm).

In 1984, H. S. White, G. P. Kittlesen, and M. S. Wrighton reported the establishment of an electrochemical transistor made on the basis of an organic polymer film brought in contact with an electrolyte [21]. The electrochemical transistor has three electrodes, two of which conduct the operating current, while the third controls magnitude of this current. Electrodes are made in the form of inert metal films. These operating electrodes are deposited on an insulating base and separated by a polymer layer. The control electrode is located above them in a thin layer of electrolyte. Conductivity of the polymer varies depending on the degree of oxidation, which is defined by the size of the potential applied to the control electrode. As a result of electrochemical oxidation or reduction, the concentration of dopants in the polymer changes and switches the resistance between the conducting, semiconducting and insulating state.

Advantages of electrochemical transistors based on organic polymers over electronic transistors are numerous. Firstly, they can operate at lower voltages (< 1 V). They can also be used as integrators and elements of short-term memory because they remain in the same state after the removal of voltage [22]. In addition, they can be manufactured by inkjet printing technology that is much simpler and cheaper than the methods of photolithography currently used in the manufacture of integrated circuits in solid state electronics [23]. Manufacture of such electro-chemical transistors can be performed under normal environmental conditions, i.e. they don't need vacuum requirements necessary for electronic transistors. Applicable polymers are dissolved in the corresponding organic solvent and then printed on the insulation substrates with inkjet printers. The electrochemical transistors created on the basis of organic polymers and inkjet printing technologies can be manufactured in a simultaneous cycle with other elements of printed circuit boards, such as for example, with resistors, inductors, or batteries. In particular, transistors (due to their multifunctionality) allow one to create a complete set of all elements of circuits placed on the printed boards.

In the future, the use of equipment for bulk printing (3D printers) will allow for creation of monolithic integrated circuits on multilayered printed boards and even finished technical devices. Considering that the programs for production of these devices can be created by computers, it opens a way to creation of self-replicating and self-learning systems of artificial intelligence.

The kinematic model of a self-reproducing automaton was first proposed in the middle of the last century by John von Neumann [24]. However, so far there is no real technology for implementation of this idea. From the stated, it follows that electrochemical transistors can become a basis of such technology.

4. Electrochemical Transducers as a Part of Artificial Organs of Smell and Taste

The input of information into artificial systems is fundamental for intellectual actions. In biological systems, the receivers of information are the sense organs. Traditionally, there are five types of human senses. Vision and hearing are the receivers of information which is transmitted in the form of waves. Touch perceives thermal and mechanical information. The senses of smell and taste are based on chemical processes and are, in their natural mechanism, electrochemical analyzers of information. In the wild, taste is detected through a combination of signals from receptors in the nose, tongue and other organs. A similar principle has been proposed for creation of artificial devices called the electronic nose and electronic tongue. Both of these devices can be considered as a necessary addition to various systems of artificial intelligence.

Technical predecessors of the artificial nose and tongue are well known methods of analysis of various gases and solutions. Currently, at least ten methods of gas analysis are widely applied in industry. The highest accuracy is achieved by isotopic and chromatographic methods, in which the minimum detectable concentration is 10^{-7} percent. Using the electrochemical method, gas is first dissolved in an electrolyte and then defined by potentiometric, conductometric, amperometric or polarographic measurements. The electrochemical method is second in terms of accuracy (10^{-6} percent), but it has the advantage in simplicity of execution. However, for creation of the device modeling a nose, it is also necessary to fulfill the requirement of selective sensitivity, which in biological systems is solved by a large number of different receptors.

The human nose possesses a staggering amount of sensitivity. It can distinguish the smell of a substance comprising an amount of several tens of molecules in a mixture of several thousand substances. Furthermore, it is known that some animals have more sensitivity than even humans. However, the human nose possesses the largest genome among all mammals (over 1000 genes), providing sensitivity to smells. Therefore, when modeling the human nose it is necessary, in addition to accuracy, to fulfill at least two conditions: first, to have many different sensor inputs, and secondly, to accurately process the data received from these sensors, just as it occurs in the neural networks of the brain. These conditions were first implemented in the work of K. Persaud and G. Dodd [25].

The electronic nose can be combined along with electrochemical sensors of other types (for example, chromatographic, piezoelectric, thermo-catalytic) capable of recognizing molecules in the gas state. At present, progress in development of an artificial nose device is being advanced by the appearance of new opportunities in the field of nanotechnology. In [26] there is report about the development of a multisensory element on the basis of the field transistor in the form of carbon nanotubes. In this device, a protein receptor layer is coated on the surface of each nanotube: the interaction between the protein and the analyzed component in a gas mixture changes the current value flowing through the transistor. Every crystal holds a large number of transistors, each of which, depending on the type of the protein applied onto it, is sensitive to a certain component of a gas mixture. As a result of the interaction between the protein and an analyzed component of a gas mixture, the value of the current passing through the transistor changes.

Publications about creating devices that simulate the functions of the tongue appeared in print 14 years after articles appeared on creation of an electronic nose. They were caused by the needs of the pharmaceutical and food industries. The first "electronic tongue" device was proposed in 1998 [27]. It contained eight sensors, possessing cross sensitivity to several components with different tastes. Devices of this type are intended for measurements in solutions, and therefore, unlike the previously developed electronic nose devices, are constructed mainly on electrochemical methods. From the very beginning, their development was based upon knowledge gained by potentiometric measurements of glass and ion-selective electrodes. In addition, during the design process of the electronic tongue, principles successfully gained during the development of the electronic nose were also utilized: i.e. multisensory design and subsequent multivariate processing of entered data.

Vlasov Yu., Legin A., Rudnitskaya A. [28] together with other members of the Laboratory of Chemical Sensors at the University of St. Petersburg published a number of the fundamental works and theories which allowed for the creation and further development of more advanced electronic tongue designs. They proposed an empiric method for estimating cross-sensitivity and developed a flow-injection version of the electronic tongue that allows it to carry out multiple and repetitive measurements in automatic mode. Several hundred membrane materials for various sensors were investigated, including chalcogenide glasses, plasticized polymers containing active agents, and polycrystalline composites. The main principle behind their "massif of potentiometric sensors" was the serial measurement of the EMF of multiple electrochemical cells, each of which triggered a single sensor. Control of the measurement procedure was carried out by a computer. Methods of pattern recognition and multivariate calibration were used for interpretation of the received information. When aprioristic information about measured samples was absent, classification was carried out by means of self-learning methods.

In addition, in the above mentioned studies it is reported that the electronic tongue can identify multi-component liquids by a method similar to fingerprinting. This ability opens a new approach to the analysis of quality of many products and the determination of their compliance with the specified product taste.

From the considered results of development and research of devices for definition of smell and taste, it is apparent that their improvement occurs on the basis of application of electrochemical sensors. Processing of received multisensory information is carried out by methods of self-learning and pattern recognition by artificial neural networks. Furthermore, in sections 2 and 3 of this review it was demonstrated how essential progress in the field of creation of artificial neural networks and computing systems can be reached: this can be achieved by electrochemical information transducers such as memristors or transistors, which are based on organic polymers. These transducers allow the potential to carry out the most important functions of artificial intelligence, namely self-learning, self-improvement and even self-replication. As a result, they open a way to creation of intellectual systems with elements that will operate, mainly, on the basis of electrochemical mechanisms.

5. Conclusion

Electrochemical transducers of information developed thus far allow important advantages for technical implementation within various systems of artificial intelligence. The application of memristors and other similar electrochemical elements allow the creation of computers with new functionality, more compact layout of basic structures, longer retention time of stored information, data storage without consumption of energy, and higher ability for self-learning and pattern recognition. Electrochemical transistors based on organic polymers simplify manufacturing technology for a number of computer devices, allow them to operate at lower voltages, create the possibility of production by methods of 3D inkjet printing technology, and thus open the way for the creation of self-learning and self-replicating systems.

The considered trend in development of artificial intelligence systems brings their structure closer to the principles of organic intellectual systems founded on electrochemical and chemical mechanisms established in nature.

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