Technology and application of electrochemical motion sensors

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Abstract

In this paper, we consider technology and application of electrochemical transducers (ECT). Electrochemical systems are very promising for the development of a new element base for microelectronics. One of the most important directions is the design of micro-sized electrochemical acceleration and pressure sensors, and one of the most promising technologies is electrochemical transfer. Electrochemical transducers are very sensitive and energy-efficient and best suited for measuring weak mechanical movements. The basic principle consist in the following: microelectrodes are formed on a silicon chip and the chip is placed in a container with liquid electrolyte. Under the influence of an external mechanical signal, the electrolyte liquid starts moving (by inertia or by pressure) and transfers ions between the electrodes. The generated electric current is proportional to the external mechanical signal. We have developed a new microelectronic technology for electrochemical transducers, and have designed new instruments based on new electrochemical microelectronic chips. Currently, ECTs are applied for a variety of engineering tasks. ECTs are used to record the ground oscillation in the railway, in the tsunami early warning system, in electrochemical hydrophones for sea-bottom stations and for geoexploration systems, in the precise azimuth determination inertial systems. Copyright © VBRI Press.

Keywords: Planar electrochemical systems, electrochemical transducer, microelectronic technology, mass and charge transfer.

Introduction and historical review

At present, there are many types of electrochemical detectors and sensors working on the principles of molecular electronics. The principles of the devices operation are diverse: the concentration polarization of electrodes, electrokinetic phenomena, anodic dissolution (or cathodic electrodeposition), convection of a nonuniform electrolyte in the gravitational field, and some others. More details about various ways of converting mechanical (and other) signals in electrochemical systems can be found in [**1-4**].

Instruments based on molecular-electronic transfer (MET) are widely used in devices for measuring mechanical signals. One of the fundamental works investigating the possibility of creating molecular electronic transducer motion parameters belongs to Larkam [**5**] and dates as back as 1965. Around the same time, the first workable samples of molecular-electronic transducers (MET) were created in the Sevastopol Branch of the All-Union Scientific Research Institute of Current Sources [**1-3**].

In the 1970s, a number of modifications of diffusion sensors and devices based on ECTs were developed in the USSR. These were borehole microbarographs and seismometers, high-precision low-frequency sensors for angular and linear accelerations, sensitive sonar and acoustic sensors, compact tilt sensors. In the USSR, molecular-electronic transducers were produced serially in the Electronic Radio Equipment Plant in Sevastopol (ERA). These devices were used to record the earthquakes and air and underground nuclear explosions, including those on the other hemisphere of the earth. After the collapse of the Soviet Union, ERA plant came within the jurisdiction of Ukraine. After a while, the plant turned bankrupt and the investigations of electrochemical sensors were terminated.

In the USA, electrochemical sensors were developed in the interest of US-Navy [**6-8**]. They were used in low-frequency acoustic wave detectors, infrasonic microphones and underwater pressure sensors [**9, 10**]. Later, the development was stopped.

Scientific researches of electrochemial transducers (ECT) were resumed in MIPT at the end of the last century under the guidance of Professor V.A. Kozlov. His review of research and development of electrochemical devices is given in [**11**], where a number of specific implementations of devices based on the principles of molecular electronics are considered. The review [**12**] testifies to a significant progress over the past years.

First of all, ECTs were used in the development of Russian high-precision seismometers [**13-16**], bottom seismic stations [**17-20**], and compact instruments for seismic exploration [**21, 22**].

To date, a large number of original experimental instruments and systems have been developed based on the MET technology for different applications. For example, a device for precise azimuth determination to geometric north [**23, 24**], a system of personal inertial navigation of a pedestrian [**25**], a system to monitor the oscillations of high-rise buildings [**26-27**], a seismic guard system to register the trajectory of the "offender" [**28**], instrument to measure of rotational vibrations of the ground surface for remote monitoring of the drilling process [**29**].

Main principles and major features of the electrochemical transducers

The scheme of the planar transducing structure (cross section) is shown in **Fig. 1**. Various binary electrolytes can be used in the transducer to provide reversible oxidation-reduction reactions, for example: iodine-iodide, ferri-ferrocyanide, etc. Electrodes are made of a metal, which only involved in the electron exchange, but does not participate in the exchange of cations. This provides a theoretically infinitely long operation time of the device without changing the operating parameters.

Currently, iodine-iodide systems with platinum electrodes are most widely used. The electrolyte consists of a highly concentrated aqueous solution of potassium iodide KI (the lower limit of the temperature range is -15 °C) or lithium iodide LiI (the lower limit of the temperature range is - 55 °C) and a small amount of molecular iodine I_2. With iodide in excess, iodine forms a well-soluble complex compound "triiodide" according to the following scheme:

$$I_2 + I^- \rightarrow I_3^- \quad (1)$$

When the current passes through the MET the following electrochemical reactions occur at the electrodes:

- reduction of iodine at the cathode:

$$I_3^- + 2e \rightarrow 3I^- ; \quad (2)$$

- oxidation of iodine at the anode:

$$3I^- - 2e \rightarrow I_3^- . \quad (3)$$

The potassium ions play the role of a background electrolyte and do not participate in the reactions at the electrodes.

The operation of the transducer is based on the fact that the current through the transducer is largely determined by hydrodynamic motion of the solution caused by external mechanical disturbances. The rate of chemical reaction at the electrodes is much greater than the velocity of reactants delivery to them. When the reaction proceeds, a concentration gradient of the reacting ions is established, and the charge in the electrolyte is transferred by molecular diffusion from one electrode to another. If the fluid comes into motion under the influence of inertia forces, a convective ion transfer occurs along with molecular diffusion, which dramatically changes the velocity of reacting substances delivery to the electrodes. Accordingly, the current through the electrodes changes, as shown in **Fig. 2**. (Here and in the following figures, the letters "A" and "C" denote the anodes and cathodes respectively). The linearity of the output signal by acceleration can be provided in a wide frequency and dynamic range.

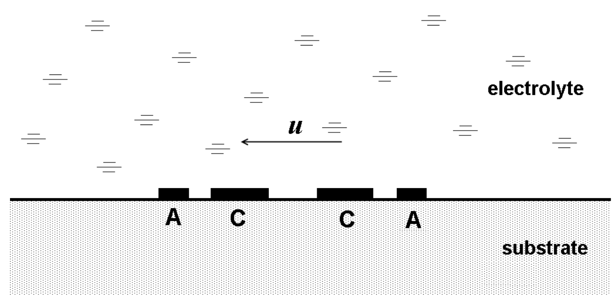

Fig. 1. Schematic representation of the transdusing electrode structure (cross section), ***u*** is the flow rate of electrolyte.

The principle of MET operation can be easily explained using the approach of flat electrodes, permeable for liquid, but impermeable for charge [**5**].

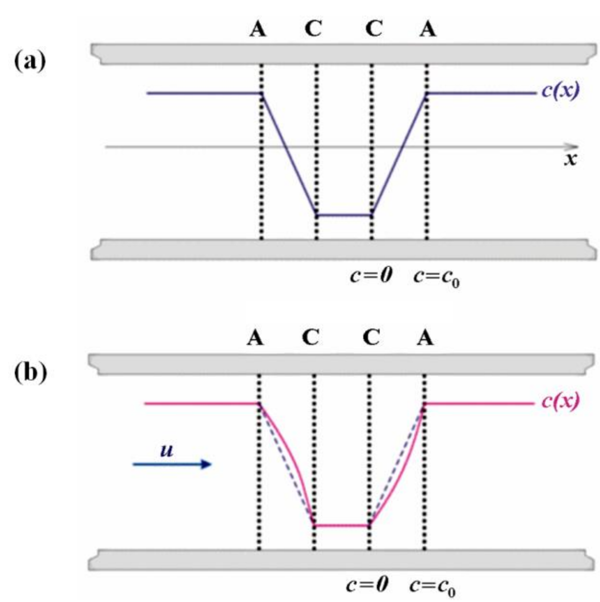

Fig. 2. Distribution of electrolyte concentration in the transducer with flat semipermeable electrodes. c_0 is the equilibrium concentration of electrolyte; c(x) is the concentration, established under a bias voltage; In the lower figure, the concentration distribution c(x) is changed by the flow of liquid.

When an electrical voltage is applied, the electrochemical current proceeds ("background current") independent of the presence of mechanical motion (**Fig. 2a**). In this case, the electrochemical reactions create a concentration gradient of the solution components, and charge in the stationary electrolyte is transferred by ions diffusion from one electrode to another.

An external mechanical signal causes the electrolyte to move under the influence of inertia forces, and to bring additional ions. An additional electric current arises in the system (along with the "background"), which is proportional to the external mechanical signal.

In other words (**Fig. 2b**), the fluid flow resulting from mechanical motion distorts the stationary distribution of carrier concentration in the interelectrode space, which leads to a strong change in the concentration gradient near the electrode surface. The dependence of electric current through the electrode on the concentration gradient near the electrode surface is described as follows:

$$I = -Dq \oint_{S} \left(\nabla c \cdot d\mathbf{s}\right) \qquad (4)$$

where D — diffusion coefficient; q – charge transferred by one ion, the integration is over the electrode area S. This system is very sensitive to a mechanical signal. Types of sensors based on ECT are shown in **Fig. 3**.

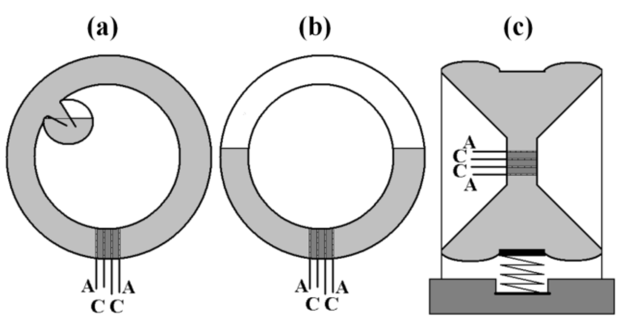

Fig. 3. Types of sensors. (a) rotational; (b) horizontal; (c) vertical.

The main advantages of devices based on ECTs

- High efficiency of converting a mechanical signal into electrical current at the level of physical processes occurring in ECT;
- A low power consumption;
- Short time to operating mode;
- The sensors do not contain parts of precision mechanics;
- Relatively inexpensive and easy to manufacture;
- ECTs-based devices are easy to operate.

Comparison of ECTs and precision mechanical seismometers

Expensive precision electromechanical and magneto mechanical seismometers (Guralp, Streckeisen, Trillium), have a number of disadvantages, despite excellent characteristics. The most significant drawback of mechanical seismometers is the complexity of the mechanical system and the very high demands on the quality of precision mechanics manufacture, which makes them very expensive. Among other disadvantages are significant restrictions on the devices transportation, large weight, high energy consumption and a long time to enter the operating mode.

Seismometers based on ECT successfully compete with those, because they are of significantly smaller weight and cost. Electrochemical seismometers are undemanding to the conditions of transportation, quickly go into the operating mode, have a low power consumption, and are easy to operate.

Comparison of ECTs and fiber optic devices

With the development of quantum optics, fiber optic sensors are becoming increasingly widespread. At comparable characteristics, the main advantages of ECT are their low price and the ease of manufacturing.

Comparison of ECTs and MEMS

High-quality MEMS, simple and inexpensive, have recently appeared. Their operating frequency range is in the higher-frequency spectral region. In the region of low and infra-low frequencies (from 0.01 Hz to 10 Hz) MEMS characteristics are unsatisfactory.

ECTs low-frequency characteristics are 100-1000 times better than the parameters of all known micromechanical devices, first of all, in terms of the self-noise level.

Comparison of ECTs and microelectronic piezoelectric sensors

Piezosensors can operate in the passive mode, without power supply, while ECTs require a reference voltage of 300-500 mV. However, expensive high-precision piezoelectric sensors require power supply. ECTs low-frequency characteristics are also better than the parameters of the best modern piezosensors. On the other hand, piezosensors capable of operating in a wider pressure range, up to 1000 atmospheres.

The limitations and disadvantages

Modern electrochemical seismometers are slightly inferior to the best models of precision electro- and magneto mechanical seismometers in terms of self-noise in the high-frequency region.

When compared with MEMS, the upper limiting frequency (not higher than 3 kHz to date, which is typical of the majority of relatively slow modern electrochemical systems) should be attributed to ECTs disadvantages, and also, higher cost.

The limitations of the temperature operation range are connected with boiling and freezing temperatures of the electrolyte. The temperature range with low-temperature electrolytes is -50°C ... + 65°C (-58°F ... +150°F).

Modeling and design of devices

Many articles were devoted to the study of physical processes in the molecular-electronic transducers [**30-36**]. Various electrode systems were studied (**Fig. 4**).

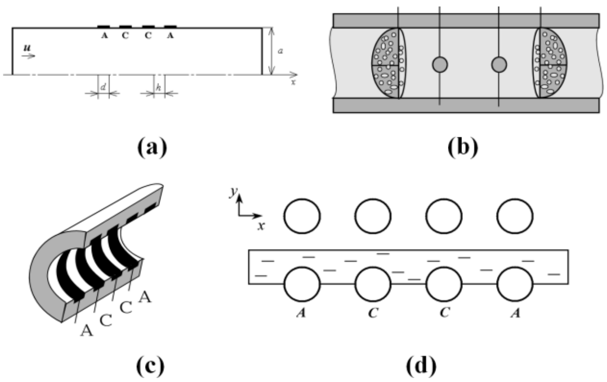

Fig. 4. Studied transducers with various electrodes: (a) Flat channel with electrodes on the inner surface [**30, 31**]. (b) Transducer with a spherical electrode structure [**32, 33**]. (c) Cylindrical channel with electrodes [**34**]. (d) Lattice of round cylindrical electrodes [**35, 36**].

It was established that the most important parameter of the transforming electrode structure is the distance between the cathode and the anode. Reducing this distance leads to an increase in the upper operating frequency, to a reduction of the nonlinear distortion and to an expansion of the operating dynamic range. Diminishing the electrode and channel size reduces the level of the transducer intrinsic noise. In particular, this suppresses the mechanism of the nonequilibrium hydrodynamic noise associated with the convective instability in the liquid in the converting element [**37**].

A worldwide interest exist to new electrochemical sensors. Several scientific groups develop and test planar structures executed at a high technological level. One of the groups [**38-41**] makes multi-crystal devices with bilateral metallization (**Fig. 5**, **Fig. 6**).

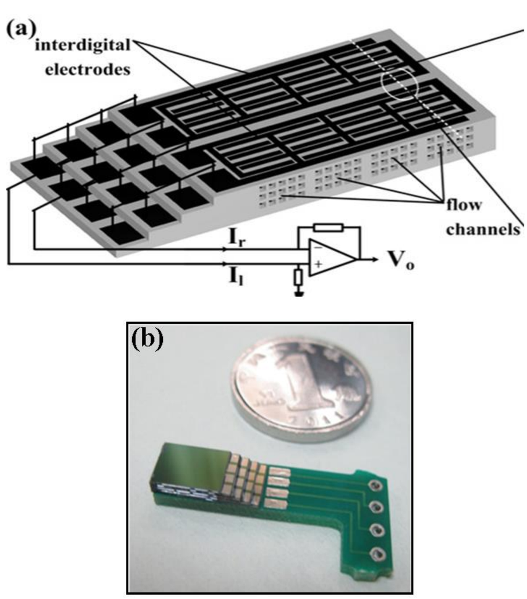

Fig. 5. Multichannel transducer. (a) Schematic of the micro device, (b) multichannel microdevice sample [**38, 39**].

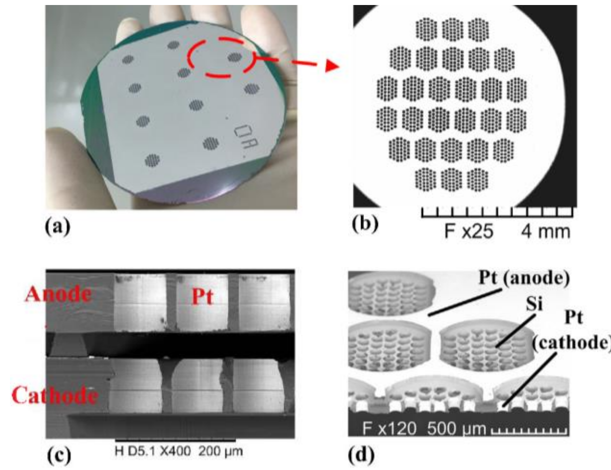

Fig. 6. Two-crystal sample with bilateral metallization. (a) Silicon wafer after fabrication; (b) SEM image of the cross section; (c) SEM image of the cross section; (d) Scanning electronic microscopic (SEM) image of the anode (the front side) [**40, 41**].

Another group is developing devices printed with ceramic pastes of different conductivity. A 3D-printed metal-ceramic element is shown in **Fig. 7** [**42, 43**].

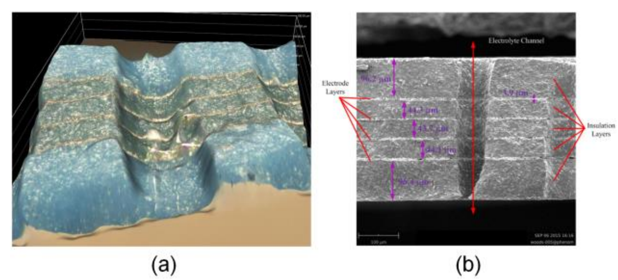

Fig. 7. 3D-printed metal-ceramic sensing element with microflow channels and dimensions of the intersection of the surface of a single channel [**42, 43**].

A very interesting trend is to use compressed microporous electrodes in electrochemical sensors, **Fig. 8**. [**44-46**].

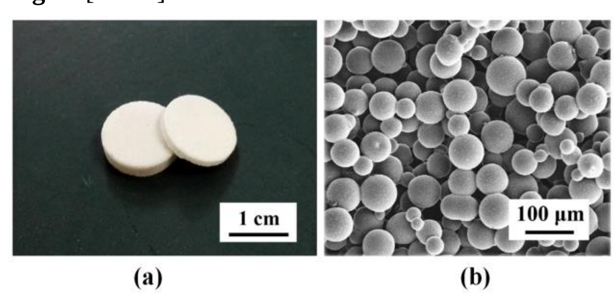

Fig. 8. Porous transducers. (a) Appearance of transducers. (b) Micro structure of transducer [**44-46**].

Microelectronic technology for electrochemical microchips

The planar electrode system has many advantages over other electrode configurations considered here. Development of a technology for industrial production of electrochemical transducers using standard microelectronic techniques would minimize the spread of parameters, reduce the power consumption and the device cost.

The works [**47, 48**] showed the technological possibility of manufacturing a microelectronic planar electrochemical transducer. The technological foundations for the production of planar electrochemical transducers are presented in [**49-51**]. The technological process is a set of standard technological operations typical of semiconductor microelectronics industry. However, the experience of using these methods for manufacturing converters of motion parameters based on the principles of mass and charge transfer in electrochemical microsystems has been very small. We developed and fabricated a number of samples, shown in **Fig. 9**.

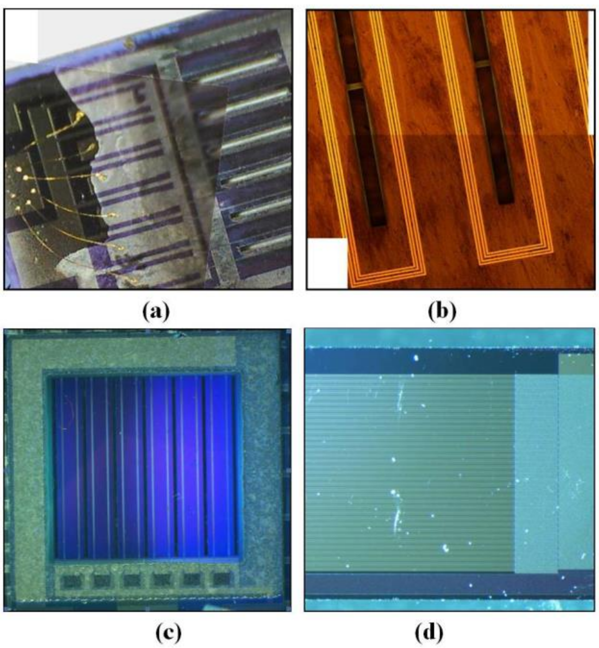

Fig. 9. Our samples of electrochemical transducers. (a) Experimental single-layer sample with welded interlinks, IMT RAS; (b) Experimental single-layer sample, MIPT in cooperation with the Technological Centre MIET; (c) Pre-series multilayer sample developed with Mikron plant & JCS MERI; (d) Perspective sample with a high electrode density developed with JCS MERI.

The technology development begins with modeling the planar structure. We simulate electrochemical processes of charge transfer in microstructures, and calculate the characteristics of the future device depending on the system parameters.

As an example, **Fig. 10** shows the distribution of the main ions concentration in a flat channel near the electrodes for different distances between the electrodes. The next step is the 3D modeling of a multi-layer microchip of the calculated thickness and dimensions. **Fig. 11** shows one of the models of our microchips. Two layers of metallization, separated by a dielectric layer, are deposited on the dielectric substrate with through holes. Then, the microchips are manufactured on 100 mm plates (**Fig. 12**). The crystals are cut and packed in ceramic housings, which are resistant to the reactive electrolyte (**Fig. 13**). The manufactured microchip is further used in devices.

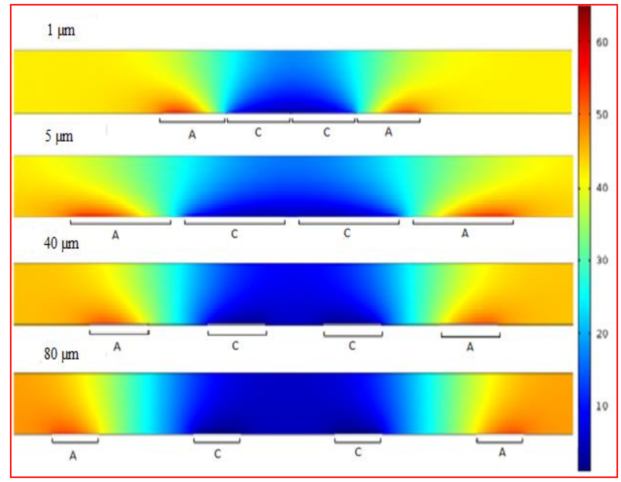

Fig. 10. Modeling and simulation of the electrochemical transfer. The distribution of the main ions concentration in a flat slit channel near the electrodes is calculated at different distances between the electrodes.

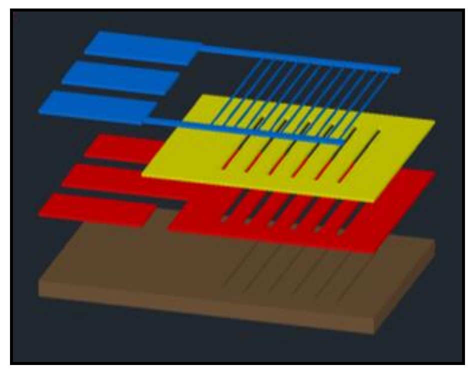

Fig. 11. Chip design and development. Multilayer structure of electrodes. Two layers of metallization (red & blue), separated by a dielectric layer (yellow), are deposited on the dielectric substrate (brown) with through holes.

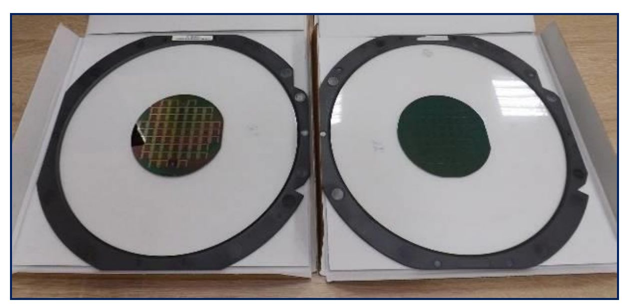

Fig. 12. Silicon wafers with electrochemical microchips.

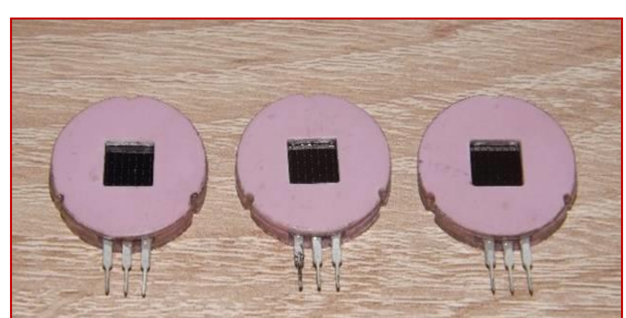

Fig. 13. Microtransducers in a ceramic package.

Devices based on the electrochemical planar transducers

We have developed several devices for different purposes, based on the new electrochemical chip. A geophone for exploration geophysics, a precision accelerometer and an ultra-low frequency hydrophone are shown on **Fig. 14**. All of them have battery-powered modifications. Their brief characteristics are given in **Table 1**.

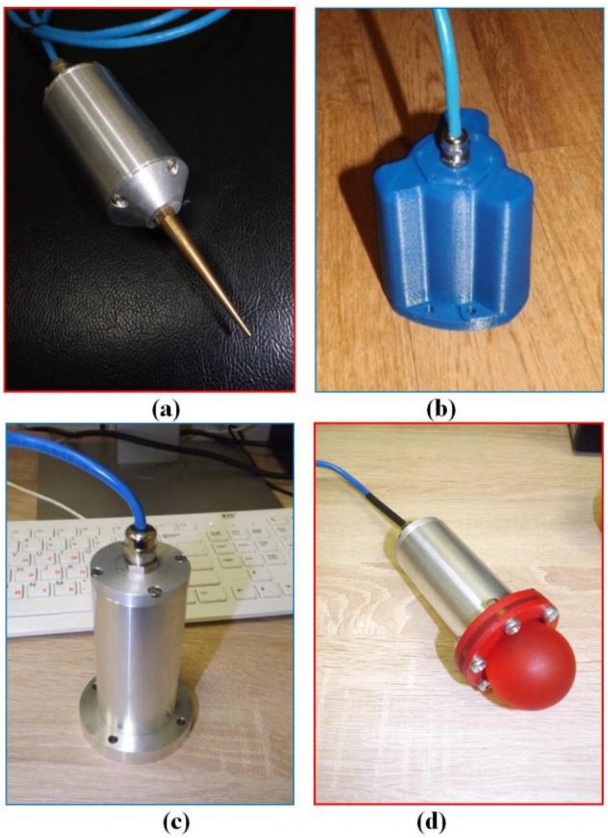

Fig. 14. Examples of applications: (a) Geophone for exploration geophysics; (b, c) Precision accelerometers; (d) Ultra-low frequency hydrophone.

Table 1. Characteristics of accelerometer, geophon, hydrophon.

	Accelerometer	**Geophone**	**Hydrophone**
Frequency range	0.1 - 300 Hz	0.1 - 300 Hz	1 - 100 Hz
Sensitivity	6.0 V/g	100 V/(m/s)	1mV/Pa
Dynamic range	115 dB	100 dB	100 dB
Intrinsic Noise	110 ng/√Hz	300 nm/s	10 µPa

For the experiment, we designed a modular system with modules for various purposes in standardized enclosures (**Fig. 15**). It allows assembling a flexible system with the necessary characteristics from individual modules like children cubes.

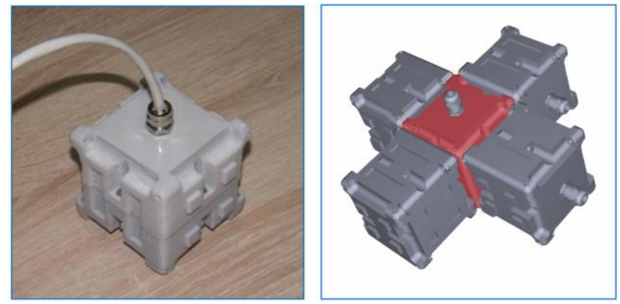

Fig. 15. Examples of applications: universal measurement unit module, and a measuring system assembled from modules for different purposes.

Conclusion

The technology and application of electrochemical transducers (ECT) are considered. Electrochemical systems are shown to be very promising for the development of a new element base for microelectronics. Electrochemical transducers are very sensitive and energy-efficient and best suited for measuring weak mechanical movements.

The planar microelectronic technology is the most technologically advanced method of manufacturing of electrochemical transducers. Planar electrochemical transducers can be used in accelerometers, seismic sensors, rotation sensors, hydrophones and pressure sensors.

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Supporting information

Supporting informations are available from VBRI Press.

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