

On precise pH and dpH measurements

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Abstract—In this paper we investigate the long-term dynamics of pH and ORP measurements in organic and inorganic liquids subjected to a modulated 'high-penetrating emission' of different nature. Development, calibration and tests of highly sensitive differential pH -meter are shown. Performed experiments aim to explore the effect of information imprinting appeared by a modulation of natural and artificial sources of emission. This methodology and measurement devices represent an experimental method for laboratory analysis of weak and ultra-weak effects in complementary medicine and 'information pharmacology'.

I. INTRODUCTION

As shown by independent studies, see [1], [2], [3], [4], [5], the 'high-penetrating emission', produced by different generators, changes physicochemical properties of liquids. For example, [6], [7] reported on impacting the dielectric properties of water by such emission. The performed experimental verification by conductometric methods confirmed this effect, however several limitations imposed on the measurement method were discovered [8]. Thus, measurements of water conductivity by alternating current, utilized in most industrial conductometers [9], [10], do not lead to a positive result. However, these changes were confirmed when performing a long-term high-resolution measurement by DC current. It is assumed that a deep polarization of electrodes and structural changes of water dipoles in so-called electric double layers [11] are responsible for this effect. In subsequent papers [5], [12] it was shown that a high-frequency conductometry also detects changes in conductivity of liquids exposed by the 'high-penetration emission', which, in this case points to a spatial polarization of dipoles in the volume of fluid.

Several authors, e.g. [13], [14], [15], pointed to changes of other parameters, such as pH and oxidation-reduction potential (ORP). We have also reported on the possibility of detecting the 'high-penetrating emission' by potentiometric methods, in particular by EMF created by bimetallic electrodes [16]. Since these measurements are parts of classical physicochemical analysis, it was decided to replicate these pH and ORP measurements. The main goal was to develop a fast potentiometric method for detecting

this emission in field conditions by using commercially available devices.

In the performed experiments, a LED generator was first selected as a source of such 'high-penetrating emission'. Its ability to generate this emission has been demonstrated in previous experiments [8], [17], [16], [5], also including long- and super long-range experiments [18]. Capabilities to imprint water and small biological objects (such as seeds) by modulating the emission of this generator is also recently demonstrated by a large number of statistically significant results [19]. Several experiments were conducted with so-called passive generators based on the shape (geometry) effects [20]. Finally, we also performed a number of experiments with 'information copies of biologically active substances' – the technology *IC Medicals*¹ [21].

It has been observed that so-called 'activated' objects, i.e. subjected to a 'modulated' emission, cause different response from measuring devices than 'non-activated' objects. For example, two identical objects placed in a differential pH -meter (dpH -meter) do not cause any response (or only a weak reaction). If one of the objects is 'activated', it causes a significant response by amplitude and phase of signals. Thus, by using a precision dpH -meter, which represents a classic potentiometric method of physicochemical analysis, it was possible to measure the effects of information imprinting, both qualitatively and quantitatively. Despite the imprinting effect being discussed in a controversial manner by many communities, the precision dpH approach allows the conduction of objective measurements in such areas as homeopathy, 'information pharmacology' or 'bioresonance therapy' [22] by a widely recognized method.

Preliminary experiments with commercially available devices indicated that they are not suitable for low-signal pH measurements due to electronic noise and low sensitivity. It has been assumed that, alike the case of conductometric approach, such devices are not designed for performing measurements with required precision, both in terms of sensitivity and resolution of the device and the long-term thermal stabilization of samples. For example, the resolution of most pH meters is in the range of 0.01 pH , the repeatability of measurements – ± 0.02 pH . The experiments however showed that changes caused by a modulated 'high penetrating emission' are in most cases

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TABLE I
MEASUREMENTS OF ORP AND pH OF DIFFERENT FLUIDS (1) BEFORE, (2) IMMEDIATELY AFTER AND (3) 8 HOURS AFTER THE
ESPLOSURE BY A LED GENERATOR. THE ORP VALUES ARE SHOWN WITHOUT CORRECTION (207mV FOR 25°).

N	F1		F2		F3		F4		F5		F6	
	mV	pH	mV	pH	mV	pH	mV	pH	mV	pH	mV	pH
1	213	7.70	149	7.68	60	6.82	137	3.53	84	8.37	186	2.77
	175	7.71	186	7.75	63	6.83	154	3.52	103	8.39	170	2.79
	179	7.71	156	7.74	53	6.80	155	3.52	82	8.44	172	2.79
2	238	7.71	185	8.16	41	6.82	161	3.52	133	8.30	171	2.79
	234	7.70	147	8.15	41	6.82	175	3.51	124	8.31	172	2.78
	231	7.71	130	8.15	37	6.81	174	3.53	80	8.35	170	2.79
	242											
3	242	7.40	180	8.16	66	6.78	174	3.49	153	8.49	173	2.79
	267	7.41	134	8.17	60	6.79	160	3.50	100	8.50	170	2.79

at 10^{-3} - 10^{-4} pH for 10 hours or more. Moreover, nonlinearities caused by a long-term unstable temperature make impossible a reliable detection of such weak and ultra-weak impacts without thermostatic functionality of existing pH /ORP devices.

After these preliminary experiments, the requirements for a new measuring instrument and methodology were formulated. They relate to a high resolution of the measurement channel (up to 10^{-6} - 10^{-7} relative pH), ultra-low electronic noise (at $< 1\mu V$), the possibility of long-term battery operation (weeks), long-term thermal stabilization of samples (at 0.01-0.005 °C relative temperature). In this work we show the development, calibration and tests of such highly sensitive dpH -meter. This work does not aim at demonstrating significant results of pH and dpH measurements for different types of impacts – this is a task for future works.

This article has the following structure. Section II shows the preliminary experiments with industrial devices. Section III is dedicated to the development of precise dpH meter, and Section IV describes experiments with this device. Section V draws some conclusions from this work.

II. PRELIMINARY MEASUREMENTS WITH COMMERCIALLY AVAILABLE DEVICES

To estimate the possibility of using potentiometric methods for detecting/characterizing the 'high-penetrating emission', we first conducted preliminary tests with commercially available instruments. Tests with pH and ORP approaches were performed in 2012, experiments with EMF in water [23] by using bimetallic electrodes – in 2013 and 2014, see [16]. Conductometric experiments with fluids have been performed during 2011-2014, see [5], [8], [17]. In total, about a hundred of experiments with different industrial devices have been conducted; in this section we show only a few characteristic experiments.

For preliminary pH and ORP tests the six following liquids were selected:

- 1) F1: Distilled (bidistilled) water
- 2) F2: Drinking bottled water 'Aquarel'
- 3) F3: Milk 1.5% fat, pasteurized
- 4) F4: Wine, dry, red, 13%
- 5) F5: Vodka, 40%

- 6) F6: Nectar of black currant, 50% juice

Choice of liquids is motivated by their use in household. Fluids were poured in plastic containers measuring 50 mls each. Measurements were carried out by laboratory devices² with special electrodes for pH measurements – GE100 and ORP measurements – GE105, produced by Greisinger Electronic (GHM Messtechnik GmbH). Measurement accuracy is ± 0.02 pH , Redox – 1 mV.

pH and ORP electrodes are new, never used in other experiments. As recommended in the manual, each ORP measurement lasted about 10 minutes, after that the electrode was washed in water, in solution of hydrochloric acid and pepsin (0,1 HCl + Pepsin, the set GRP100) and finally 10 minutes soaked in distilled water. The pH electrodes were also washed in water and stored in 3M KCl solution. For each liquids at least 3 ORP and pH measurements were conducted. Each experiments took about 5 hours, the laboratory was illuminated only by artificial light.

All containers were installed in front of the generator at the distance of 30cm, the generator itself (LED, high-voltage, 470nm) was enclosed in a grounded metal container made of 1mm thick steel. The intensity of electric and magnetic fields around the system was reduced up to the level of $\pm 10nT$ and $\pm 1mV/m$, (see, e.g. [24] on the influence of magnetic field on the water). Exposure lasted 60 minutes. Several results are collected in Table I.

For further ORP measurements three inorganic liquids F1, F2 and F5 were chosen. For F2 we also measured the level of pH . In contrast to the previous measurements, this time the electrodes were in the fluid during the entire experiment. Each of these experiments lasted for about 4-5 hours: one hour before exposure to the generator, one hour of the exposure to the generator (shown as gray bar), and at least two hours after the exposure. The data was transferred to a computer every 10 seconds and automatically written into a file. The results are shown in Figure 1.

²The device name is not specified for commercial reasons. The device and the electrodes are in the price range of 'up to 500 euros', i.e. this is a high-precision measurement system designed for laboratory and general applications.

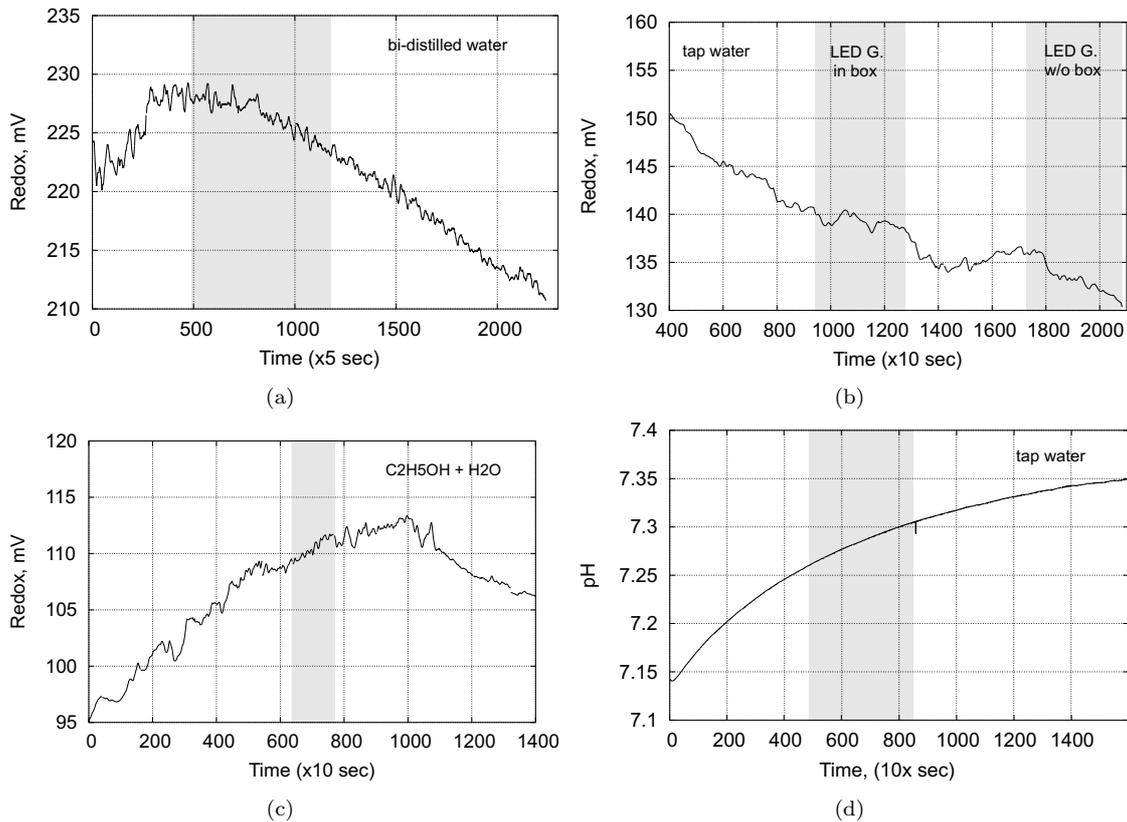


Fig. 1. (a, b, c) ORP measurements of three test liquids for 4-5 hours (before, during and after the exposure to the generator). In (b) we show two types of effects: the generator in a metal container (and without such a container); (d) graph of pH water. The gray line shows the exposure to the generator.

A. Conclusions from preliminary experiments

The main objective of preliminary experiments was to identify significant changes of pH and ORP when the fluid was exposed to the emission. However, as shown in Table I and Figure 1, none of the measured parameters indicated a significant correlation with the exposure time within a 4-5 hour period.

ORP measurement is a time-consuming process, about 5-10 minutes per measurement, which also requires a calibration and cleaning of electrodes in the buffer solutions. Measurement accuracy is low since the time required to reach a steady state differs between measurements. As observed, this time also depends on the previous liquid, e.g., the first measured values are almost always larger than other values (ORP electrodes are stored in a special fluid). ORP variation in Table I for the same liquid and the same measurement can be explained by this factor. While the device contains a built-in temperature sensor, measurements demonstrated an essential temperature dependence (this indicates the need for accurate thermal stabilization of samples).

Thus, we did not succeed to reliably measure any significant changes of ORP that correlate with exposure to the LED generator within 4-5 hours. There is a significant systematic and random error due to variations of temperature, mechanical factors, pollution of electrodes, etc. Obviously, this error is a significant obstacle to the

development of more sensitive instruments. It was decided to abandon further ORP measurements.

Measurements of pH are more stable. There were small changes of pH before and after the experiment at 0.01-0.03 pH (on a boundary of resolution). Since the temperature was not stable during this time, these fluctuations may also be caused by temperature variations (non-linearity in the electrodes, different temperature coefficients of semiconductors, etc.). Temperature compensation of the Nernst potential, which the measuring device performs, is not able to compensate these changes. Conducting long-term precise measurements, such as [13] for a few days, does not make sense without thermal stabilization.

As a general conclusion, the measurement of pH and ORP does not represent a desired express method for detecting/characterising the emission in field conditions with available commercial devices. Analysis of long-term changes (within 24-48 hours) of pH may indicate the impact of 'high-penetrating emission', but specialized instruments and new measurement approaches should be developed.

III. ACCURATE pH MEASUREMENTS

Since scientific publications (for example, works of M.Krinker [15], [14], [25], [26], [27]) and preliminary experiments have shown the possibility of using pH measurements for detecting the 'high-penetrating emission',

it was decided that it would be beneficial to develop a new specialized device. One of the motivations for a new development was the lack of affordable commercial devices for this purpose.

Development began in the fall of 2013, the first prototypes were produced late spring 2014. Basic requirements for the device are related to the resolution up to $10^{-6} - 10^{-7}$ pH , i.e. at the level of $0.5 - 0.05 \mu V$, and the measurement accuracy of $10^{-4} - 10^{-5}$ pH . This imposes strong restrictions on the noise level in the system, the quality of analog-to-digital conversion and amplification in the analog part of the device. The system should allow processing of digital signals in real time, e.g. by digital filters. Since pH electrodes have extremely high output impedance (50-500M Ω), the input circuits should have a very low input current. The system should be scalable for differential (or with the reference channel) measurements. Furthermore, device must be capable of working offline (i.e., without a connected computer) for a long time – at least several weeks. The device must also possess an accurate two-channel thermostat.

A. Description of measurement system

The measurement system is based on ARM Cortex M3 core and a set of analog modules. In particular it has 20-24 bit (with oversampling) delta-sigma ADC, whose resolution is 61nV. For pH electrodes the system has an input buffer Op Amp that provides the input current at $3 \cdot 10^{-15}$ A. The system possesses 64MB flash memory, which is enough to record all measurements (4-, 8-, 16-, 32-component vectors of measurements, each measurement is 32-bits) for a period of 60 days. For recording environmental conditions during the experiment the system has an accurate 3D magnetometer with the resolution of 80 $\mu Gauss$, 3D accelerometer with the sensitivity to linear acceleration 61 μg , internal and external temperature sensors, the Voltage sensors for power control. Other sensors can be connected by using additional analog and digital on-board modules. This set of built-in sensors is intended

to determine whether the desired effect appeared due to local environmental factors or is caused by the 'high-penetrating emission'. The whole system is assembled on a multilayer high-density PCB (for example, only FQFN chip packages are used). When the thermostat is off, the current consumption is about 5–50 mA depending on the ARM clock frequency. With a standard USB power source, the so-called PowerBank, e.g. 9A/h, the system can autonomously work about 70 days (with external temperature stabilization). The use of components with ultra-low noise and suppressing the noise in the system allowed us to keep the noise level in the measurement channels at $< 1 \mu V$. Universal measuring system received the production label 'MU' (Measurement Unit), one of the modules is shown in Figure 2.

The software part is a biggest part of the development and reflects the current trend of microprocessor-based systems to increase the software development up to 60%–70% of all developmental tasks. Measuring module uses a real time operating system. For receiving data, the module has a client program on PC, which is connected via USB interface. MU is easily scalable, e.g. by connecting N such modules via USB hub or I2C, SPI or CAN interfaces.

B. Description of the pH-meter

Two fully identical measurement channels based on the module MU2.0 have been configured for single or differential pH measurements. Due to characteristics of MU2.0, the device is able to measure the acid-base changes in the test fluids at $10^{-5} - 10^{-7}$ pH , that is not possible for most other devices. Main applications are long-term laboratory and field measurements with small and ultra-small changes of pH . The prototype of the device³ is shown in Figure 3.

This device features the possibility of a long-term battery operation (up to 60 days) without connecting a computer. The data transfer and programming of the device is performed via USB interface by using the client software on a PC. Data from embedded sensors that characterize environmental parameters – 3D magnetometer, 3D accelerometer, 4-channel temperature logger and voltage – are recorded together with the measurement data. Recording all data occurs in real time. For high-precision pH measurement device has a built-in two-channel thermostat with PID controller, which allows keeping the relative temperature at $< 0.01^\circ C$. The device can be powered from the USB interface and USB battery. For power supply and USB interface the device has 2 miniUSB connectors, D-Sub-9 for connecting 4 temperature sensors and 2 resistive heating elements for thermostat and 2 BNC connectors for pH electrodes.

Main specifications:

- number pH of channels: 2
- number of temperature channels: 4
- measurement range pH : 0-14 (4-10)

³Patents are pending.

Measurement Unit, MU 2.0



Fig. 2. One module of the measuring system MU2.0, manufactured by *Cybertronica Research*.



Fig. 3. Prototype of precision differential USB pH -meter based on MU2.0.

- resolution of measurement channels:
 - at 0-14 pH : 61nV ($\sim 10 - 6 pH$)
 - at 4-10 pH : 30nV ($\sim 5 \cdot 10 - 7 pH$)
- noise: $< 1\mu V$
- sampling frequency: 0.1s-1 hour
- resolution of temperature sensors: $\pm 0.002^\circ C$ (relative temperature)
- non-volatile memory: 64 MB
- current consumption (without thermostat): 5-50mA
- powering USB PowerBank, 9A/h: > 70 days
- dimensions (main unit) 105x70x19mm
- weight (main unit): 130g

Since the electrodes are biased with 512mV, this voltage should be subtracted from pH data (see further pH graphs).

C. Calibration of the system

The dependence of pH potential between glass and reference electrodes is described by the Nernst equation:

$$E = E^0 + 2.3 \frac{RT}{F} \log a_{H^+}, \quad (1)$$

where T is the temperature in Kelvin, R – the gas constant, F – the Faraday constant, E^0 – the standard potential at $a_{H^+} = 1 \text{ mol/L}$ [28], [29]. The value $E_N = 2.3 \frac{RT}{F}$ is known as the Nernst potential and determines the dependence between the potential and the pH value. As it follows from the equation (1), E_N depends on the temperature, thus each pH measurements must be accompanied by temperature measurements.

The need for calibration is caused by several factors, primarily by different coefficients E_N for different electrodes (even from the same manufacturer). These differences are defined by technological factors, and aging/contamination of electrodes. To estimate the deviation of real E_{Nm}

from theoretical E_{Nc} , the dimensionless coefficient K is introduced:

$$K = \frac{E_{Nm}}{E_{Nc}}, \quad (2)$$

where $K = 1$ in ideal case. Since a technological bias in electrodes is linear (or close to linear) for reaching $K = 1$ the coefficients α and β , based on measurements in the buffer (test) liquids, are calculated

$$\tilde{x} = x(1 + \alpha) + \beta, \quad (3)$$

where x is the measured value of pH and \tilde{x} is a calibrated value. To determine the α and β , two points measurement (at $pH = 3$ and $pH = 7$) are required, which should be measured at a fixed temperature.

Identical measurement channels used the same electronic components, but they have small differences in values, for example, the values of resistors and capacitors are usually defined with 0.1%–5% accuracy. For differential measurements there is an offset between channels, which is caused by different total length of conductors, gain's variations in each channel, the temperature coefficients of semiconductor components, etc. These differences are very small, but in total they may provide an additional deviation. Thus, it is necessary to perform a differential calibration of channels to obtain a differential '0' in the absence of impact. In this regard, we distinguish between the electrode calibration and the differential calibration.

As shown in Figure 4, the operation of the device is divided into five phases – phase 0: 'before measurements'; phase 1: 'temperature adaptation'; phase 2: 'no impact'; phase 3: 'impact' and phase 4: 'after impact'. Both types of calibration are performed in different phases. For calibration a precision voltage source, laboratory voltmeter, calibrated temperature sensor and buffer solutions $pH = 4$, $pH = 7$ or $pH = 10$ are required. All calibrations are performed by two points approach.

Calibration of electrodes in the phase 0 consists in setting the coefficients α and β in buffer solutions. For each electrode, these values are stored in non-volatile memory (i.e. each pH electrode must be numbered and stored in the database of the device). The initial calibration of temperature sensors (including calibration of Op Amp and ADC) occurs also in the phase 0. The device has an automatic calibration, where the system solves a system of linear equations to compute the α and β , users only needs to put electrodes in buffer fluids.

Differential calibration depends also on the temperature of electrodes (and the system as well), which is reached in the phase 1. Therefore the offset compensation for the channel must be made for the phase 2. Since each phase is specific for each measurement, the compensation should be performed *after measurements*. Thus, the differential measurement compares the behavior of the system in the phase 1 (no impact) and in the phase 2 (impact) – this fact should be noted. When only a differential measurement is intended, the calibration of pH electrodes in buffer solutions can be omitted.

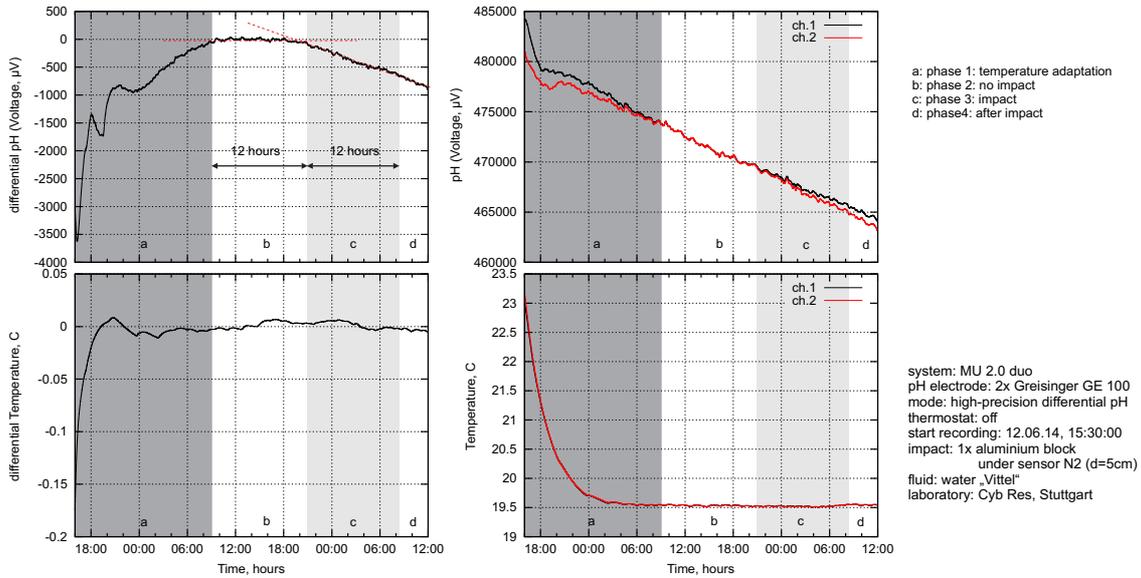


Fig. 4. Phases of measurement process. Shown is the reaction of the instrument on aluminium block under the sensor N2. The block was kept in the room for 24 hours and placed under the sensor outside the temperature oven without opening the door, i.e. no changes of temperature during the block placement (internal thermostat is turned off). Visible noise in the pH channels is due to periodic temperature oscillations in the oven (the thermostatic oven uses the threshold-based controller).

Differential calibration. Despite the differential balancing represents a linear problem, some explanations are needed for this transformation. As shown in Figure 6, data from the pH channels 1 and 2 are represented by the lines L_1 and L_2 . In the theory, the L_1 and L_2 must be strictly parallel, but in the praxis there is a difference in their slope and offset, which generates a non-zero dynamics of dpH . By a linear transformation (3) the lines must be transformed so that $dpH(L_2 - L_1) = 0$ for $t_{a-t} - t_a$.

Let us assume that the channel 2 (ch.2) is exposed. There are four different cases:

1. Dynamics in the channel 1 is not impacted by the exposure (or all deviations in this channel are ignored) and the channel balancing is not performed. In this case the change of $\Delta V'$ can only be measured in the channel 2 related to the trendline (marked by the dashed line). This strategy is used in single-channel methods [8], [16], [17]. However, it must be noted that the value of $\Delta V'$ depends not only on the intensity of exposure, but also on the slope of L_2 .

2. The dynamics of the channel 1 is not impacted, but the channel balancing is performed. In this case, one of the lines L_1 or L_2 are linearly transformed by $\alpha x + \beta$. If L_1 is transformed, then $\Delta V'$ remains unchanged. If L_2 is transformed, then the value of $\Delta V'$ needs to be scaled by $\alpha x + \beta$.

3. The dynamics of the channel 1 is also impacted by the exposure in the channel 2. This is the most typical situation. In this case, one of the channels is transformed and ΔV is considered as the difference between the data from channels 1 and 2 for $t_{a-t} + t_a$. Obviously, in this case ΔV is also scaled by $\alpha x + \beta$.

4. The dynamics of both channels are impacted by the exposure in the channel 2. However in contrast to (3), in

this case both channels are transformed. Here ΔV is scaled by both transformation coefficients.

Since L_1 and L_2 differs from each other by technological reasons, the scale factor $\alpha x + \beta$ for ΔV and $\Delta V'$ is defined by the choice of pH electrodes. This raises the question – what are the best strategies to measure the magnitude of impact independently of the choice of pH electrodes?

Since the Nernst equation (1) does not contain time, the dynamics of pH should exhibit a constant value in the steady state, which corresponds to 'almost horizontal' line⁴. Therefore, the strategy (4) makes the measurement invariant to the choice of electrodes, i.e. both lines L_1 and L_2 must be first transformed to 'almost horizontal' line and then ΔV is calculated.

For a more formal approach, we introduce a coordinate system (x,y) instead of (V,t) . Let L_1 be defined as $\alpha_1 x + \beta_1$ and L_2 – as $\alpha_2 x + \beta_2$, where

$$\alpha_1 = \frac{y_1^1 - y_2^1}{x_1 - x_2}, \beta_1 = \frac{x_1 y_2^1 - y_1^1 x_2}{x_1 - x_2}, \quad (4)$$

$$\alpha_2 = \frac{y_1^2 - y_2^2}{x_1 - x_2}, \beta_2 = \frac{x_1 y_2^2 - y_1^2 x_2}{x_1 - x_2}, \quad (5)$$

$(x_1, y_1^1), (x_1, y_1^2)$ are defined for the point t_{a-t} and $(x_2, y_2^1), (x_2, y_2^2)$ for t_a each L_1, L_2 correspondingly. We will transform L_1 and L_2 to \tilde{L}_1, \tilde{L}_2 , which go trough the points $(x_1, y_1^1), (x_2, y_1^1 + \Delta y)$. Adding a linear transformation $(\alpha x + \beta)\tilde{\alpha} + \tilde{\beta}$ to L_1 and L_2

$$(\alpha_1 x_1 + \beta_1)\tilde{\alpha}_1 + \tilde{\beta}_1 = y_1^1, \quad (6)$$

$$(\alpha_1 x_2 + \beta_1)\tilde{\alpha}_1 + \tilde{\beta}_1 = y_1^1 + \Delta y, \quad (7)$$

$$(\alpha_2 x_1 + \beta_2)\tilde{\alpha}_2 + \tilde{\beta}_2 = y_1^1, \quad (8)$$

$$(\alpha_2 x_2 + \beta_2)\tilde{\alpha}_2 + \tilde{\beta}_2 = y_1^1 + \Delta y, \quad (9)$$

⁴The transformation to the horizontal line does not make sense because it requires $\alpha = 0$, that completely nullifies the dynamics.

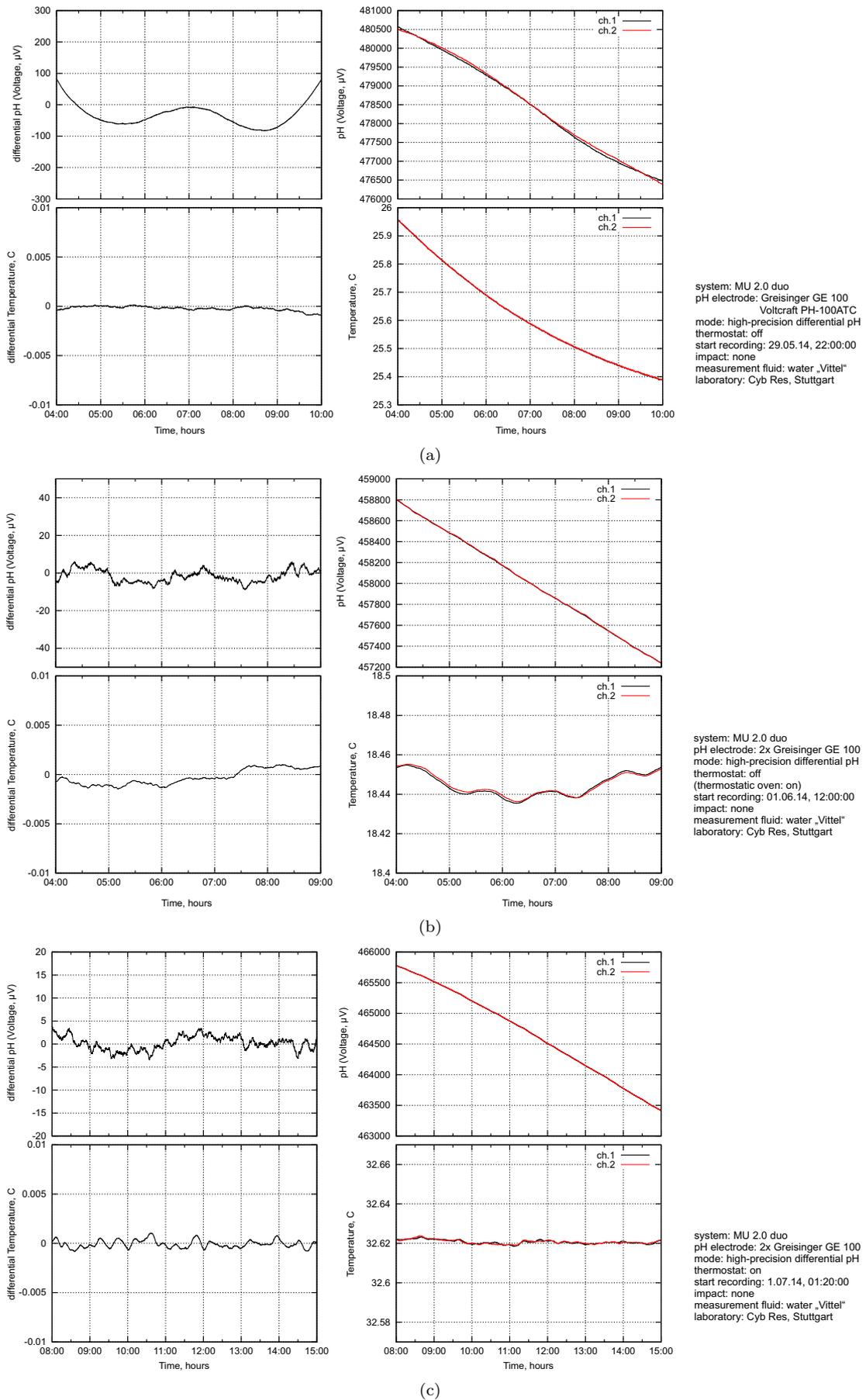


Fig. 5. Control measurements without any influences. Measurement of differential pH : (a) without temperature stabilization; (b) with external (thermostatic oven) temperature stabilization; (c) with internal (thermostat) temperature stabilization.

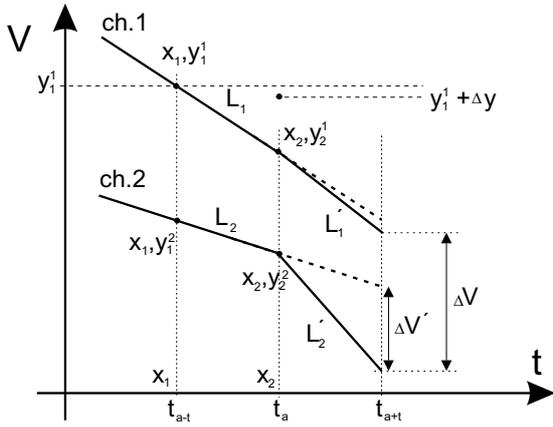


Fig. 6. Differential balancing of channels.

we receive solutions for $\tilde{\alpha}, \tilde{\beta}$

$$\tilde{\alpha}_1 = \frac{\Delta y}{y_1^2 - y_1^1}, \tilde{\beta}_1 = \frac{y_1^1(\Delta y + y_1^1 - y_1^2)}{y_1^1 - y_1^2}, \quad (10)$$

$$\tilde{\alpha}_2 = \frac{\Delta y}{y_2^2 - y_2^1}, \tilde{\beta}_2 = \frac{y_2^1 \Delta y + y_1^1 y_2^1 - y_1^1 y_2^2}{y_2^1 - y_2^2}. \quad (11)$$

Calculated coefficients $\tilde{\alpha}, \tilde{\beta}$ allow balancing the differential dynamics, i.e. to achieve $dpH(\tilde{L}_1 - \tilde{L}_2) = 0$ for $x < x_2$, or more formally

$$\begin{aligned} (\alpha_1 x + \beta_1)\tilde{\alpha}_1 + \tilde{\beta}_1 - (\alpha_2 x + \beta_2)\tilde{\alpha}_2 + \tilde{\beta}_2 &= 0, x < x_2, \\ (\alpha_1 x + \beta_1)\tilde{\alpha}_1 + \tilde{\beta}_1 - (\alpha_2 x + \beta_2)\tilde{\alpha}_2 + \tilde{\beta}_2 &\neq 0, x > x_2. \end{aligned}$$

However, as we see, for $x > x_2$ (dynamics under the impact) this transformation leads to scaled values $\tilde{L}_1 - \tilde{L}_2$ related to the initial (non-transformed) values of $L_1 - L_2$. The scaling coefficient depends on the Δy and the difference between slopes of the lines L_1, L_2 .

Practically, if pH_1 and pH_2 are the numerical values from the first and the second channel received from the device, the dpH is computed as

$$dpH = pH_1 \tilde{\alpha}_1 - pH_2 \tilde{\alpha}_2 + \tilde{\beta}_1 - \tilde{\beta}_2, \quad (12)$$

where $\tilde{\alpha}, \tilde{\beta}$ are defined by the expressions (10), (11). If both pH channel are identical, i.e. $\tilde{\beta}_1 - \tilde{\beta}_2 = 0$ and $\tilde{\alpha}_1 = \tilde{\alpha}_2 = \tilde{\alpha}$, the scaling factor is equal to $\tilde{\alpha}$ and the transformation (12) makes the measurement dpH invariant to the choice of electrodes (i.e., the different α from electrodes is compensated by different $\tilde{\alpha}$). The same situation occurs if both pH channels are not identical, i.e. $\tilde{\beta}_1 - \tilde{\beta}_2 = \Delta\tilde{\beta}$ and $\tilde{\alpha}_1 = \tilde{\alpha}_2 + \Delta\tilde{\alpha} = \tilde{\alpha}$. Here $\Delta\tilde{\beta}$ and $\Delta\tilde{\alpha}$ compensate a difference between electrodes and the $\tilde{\alpha}$ compensates a slope between different pairs of electrodes. Thus, the transformation (12) is invariant also to the choice of electrodes and, moreover, there is no need for additional scaling factors.

Temperature stabilization. Despite calibration, different temperature coefficients of electrodes and semiconductor elements introduces a nonlinearity, which manifests at temperature changes. Therefore, the essential factor is the temperature stabilization of electrodes, the system and fluid samples. The figure 5(a) shows the measurements

for 13 hours without temperature stabilization, where the variation of the absolute temperature is of 0.8°C , the differential temperature -0.05°C . The greatest impact on the system has the change of temperature gradient, which introduces a significant nonlinearity in the differential dynamics. In the worst case we observe here a 'zero drift' at the level of $300-500 \mu\text{V}$, which corresponds to $\sim 0.005 - 0.01 pH$.

Figures 5(b) and 5(c) show the pH and temperature data with external (oven) and internal (thermostat) thermostatisation. The zero drift is decreased up to $\pm 5 \mu\text{V}$ ($\sim 0.0001 pH$) and the noise to $\pm 1 \mu\text{V}$, i.e. there is an improvement of signal to noise ratio of ~ 100 . When using a dual temperature control (external temperature oven and internal thermostat) the absolute temperature stabilizes at $< 0.005^\circ\text{C}$ (with appropriate calibration of temperature sensors), which reduces the zero drift to $0.5 \mu\text{V}$, i.e. to the level of intrinsic noise in the measuring channel. In these conditions it is possible to measure the dpH at 10^{-5} .

IV. EXPERIMENTS

A. Methodology of experiments and analysis of results

The structure of experiments is shown in Figure 7(a). pH electrodes are placed in thermostats 1 and 2. There are 4 chemically identical liquids in similar containers: two measuring and two test fluids. Impact occurs on the test liquid, the measurements are made for measuring liquids. In other words there are two transmissions: 'source of exposure' \rightarrow 'test fluid' and 'test fluid' \rightarrow 'measuring liquid', which are spatially and temporary separated from each other. Test liquids are placed under the thermostats, measuring fluids are in thermostats, see Figure 7(b). These experimental conditions exclude chemical, thermal and electromagnetic transmission effects in each chain.

Instead of test liquids it is possible to use various 'activated' objects, see Figure 7(c). In this case there are also two transmissions: 'source of exposure' \rightarrow 'test object' and 'test object' \rightarrow 'measuring liquid'. Using strong emission sources instead of test objects or fluids is impractical because it substantially changes liquids in pH electrodes, and accordingly, introduces additional nonlinearities.

During analysis it needs to interpret the received data in terminology of 'high-penetrating emission'. This causes difficulties because there are no commonly accepted models for this kind of phenomena. To assist in understanding the dynamics of pH and dpH , we collected the following points:

- 1) If the test fluids were placed in one place, treated in the same manner and other similar conditions for both fluids are strictly fulfilled, we expect similar dynamics pH and as a result, very small variations in the dpH .
- 2) The test fluids may be exposed to emission from natural and artificial sources, which change their physico-chemical properties. If only one of two liquids is treated, we expect a deviation of dpH , which will characterize the degree of change in the fluid 1 in respect to the fluid 2 under the influence of emission.

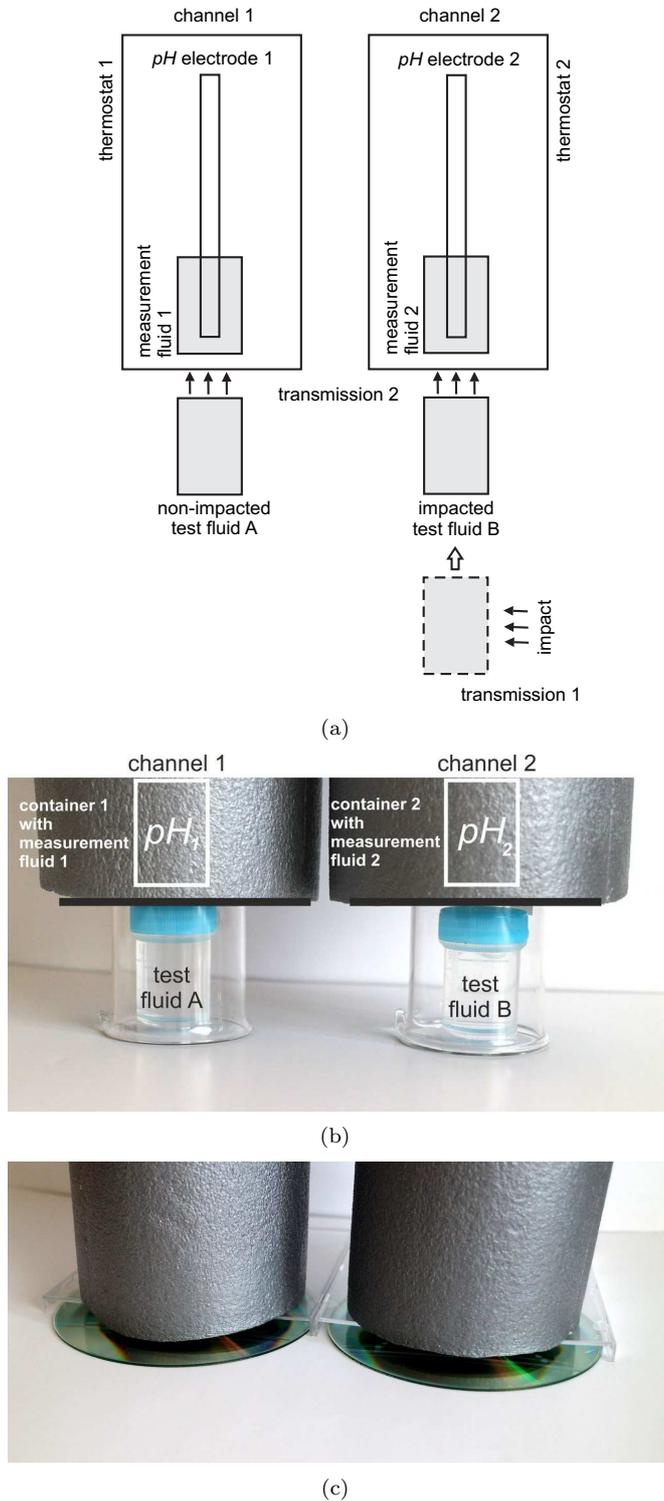


Fig. 7. (a) Structure experiments and examples of (b) test liquids and (c) test objects for control and experimental attempts.

3) Some passive objects are also sources of emission. When they are placed near the measuring liquids, we expect some changes of dpH , which are proportional to the intensity of emission. It is assumed that these changes will occur gradually over time, i.e. it is necessary to monitor a long-term dynamics of dpH .

4) There are two different factors that impact all fluids: local and nonlocal. Intensity of local factors decreases over the distance, nonlocal factors (see e.g. [18]) show to some extent no significant dependence on the distance between 'addressing objects' (nevertheless the intensity of interactions in the system 'addressing object' → 'measuring fluid' depends on the distance between them).

5) The distance, at which the local factors significantly decrease, is yet not known. Experiments demonstrated that the impact from the small objects is still perceived at distances up to 30-50 cm. If the test fluids are too close to each other, the cross-interactions between liquids and objects in both channels are possible.

6) Two identical objects located at different distances from the test fluids will impact differently on them and, as a consequence, we will observe some dynamics of dpH .

7) There is a factor of environmental influences on test/measuring/electrode liquids in form of electromagnetic fields, geobiological and other effects. So, even spatially separated fluids will exhibit different dynamics dpH , if they are exposed to different environmental influences.

8) There is an effect known as 'noising results of experiments', see e.g. [16]. It appears after a certain number of measurements: the ratio 'signal to noise' decreases, the system demonstrates various changes in the signals (even a chaotic signal) without any impact. At the moment, the way to get round this problem consists in switching off the device. After some time (usually experiment time = resting time) the device is ready for measurements again.

The experimenter has to select the experimental conditions so that at points 1), 5), 6), 7), 8) the system demonstrates close to zero (or very little) dynamics of dpH .

The analysis consists in comparing the dynamics of pH and dpH in phases 2 and 3. To do this, it needs to indicate the point t_a , i.e. to indicate when the exposure started. Since the phase 2 is linear, as shown in Figure 4, by a linear transformation (3) the obtained data need to be linearly transformed so that the dynamics in this phase is near 0. The channels are balanced for the time $t_a - t_h$, the impact is considered for the time $t_a + t_h$ (see Figure 6).

Usually, a temperature stabilization takes about 4-6 hours (with thermostats), the phase 1 – 6-12 hours, the phase 2 – 6-12 hours, i.e. the duration of one precision measurement is about 24-36 hours. Since the reference KCl fluid in the electrodes gradually mixes with measuring liquid during the measurements, the reaction time increases and the coefficient K changes. The pH electrodes need more time to reach a steady state. Therefore, it is recommended that the experiments start at least 24-36 hours before exposure (without any changes in the lab during this time), especially when pH electrodes have already been used for a long period of time. If the system loses sensitivity, it requires the replacement of the KCl liquid in electrodes or even to replace a pH electrode. Preparation time can be significantly reduced, if the measuring liquid will be not changed before each experiment and the thermostat will be not turned off. Moreover by using 2

devices, which run continuously, it is possible to reduce the measurement time up to 6-12 hours.

B. Control measurements

There are three types of performed control measurements:

- 1) without any impacts (characteristic behavior of the system in the steady state);
- 2) the same 'not-activated' objects are used in both sensors (channel equalization);
- 3) only one 'not-activated' object is used in one sensor.

Comparison between 2) and 3) gives an important information about sensitivity of this approach for common-mode and differential-mode measurements.

1. Control measurements without impacts. Control measurements of this kind were carried out as independent experiments and also performed in the phase 1 of each measurement. Examples of thermally stabilized measurements are shown in Figures 5(b) and 5(c). The behavior of the thermostabilized system without impacts is stable at $5 \mu V$ for 12 hours or more.

2. Control measurements with two identical objects. Two identical objects are: blank (and 'non-activated') CD-R discs, blocks of aluminum and containers with water, as shown in Figure 8. In the case of identical objects we observe small deviations of dpH at 50-100 μV for 12 hours. It should be noted that the reaction occurs often 2-4 hours after the start of exposure.

3. Control measurements with a single object. In these measurements only one object was placed under one (from two) sensors. The used objects are: aluminum block (see Figure 4), wood block, blank CD-R disc (Figure 10), freshly picked green leaf and the modulator 'Fortpost-1', see Figure 9. In each of these cases, the device gave a response between 450 μV and 1200 μV , i.e. there is a significant difference in measurements with one or two objects. This fact is not surprising, however, raises multiple questions about the nature of impact from passive 'non-activated' objects. At the moment, as a working hypothesis, we can assume spin-based mechanisms (or similar hypotheses), see e.g. [30].

C. Experimental measurements

Three types of control measurements showed that the differential pH -meter is sensitive to any small difference between objects. Therefore, one of the most important experimental conditions is the use of identical objects in identical positions in both channels. Since the device is stated to be capable of detecting and measuring small weak and super-weak impacts produced by 'information modulators and activators', for example, in homeopathy and 'information pharmacology', the basic idea of these experiments is to compare two identical objects in cases of 'no impact' and 'one object is activated' (another one is not 'activated'). For object activation, see e.g. [31], we used the LED generator (with information matrix) and methods of information pharmacology. Both approaches

are very similar, and both use the two-stage scheme 'generator' → 'activated object' → 'measuring liquid'. For such 'activated objects', in the case of information pharmacology, we used a CD disk, in case the LED generator, we used a container with plain water. These experiments are a continuation of works started in [8], where we also carried out the two-stage approach with the impact on test solutions and measuring the conductivity of secondary fluid.

1. The LED generator with installed information matrix. The methodology of using the LED generator with penicillin matrix for activation purposes is described in [19]. Test fluid was filled into two containers, one of them was stored close to the main bottle of water, the second one was exposed for 12 hours by the LED generator. Different modes of the LED generator are used for activation – 60 min. emission: 120 min. pause (Figure 11(a)) and 5 min. emission: 5 min. pause (Figure 11(b)). After activation and short resting, both containers are installed under the sensors, as shown in Figure 7(b). Some of the obtained data is shown in Figure 11. Physically, both water containers represent two identical objects, however when one of these fluids is activated, we observe significantly different (non-zero) dpH dynamics. Moreover, different operating modes of the LED generator reflected in different intensity of secondary emission from the test fluid. In this regard, an interesting experiment is shown in Figure 11(c). Here we used two dpH devices simultaneously, the activation was carried out in small 10 ml. containers (in the previous cases we used 50 ml containers). Both devices showed lower values (about 120-140 μV), than in the previous cases, moreover there is a significant common-mode component (probably explained by a close position of all 4 containers). It can be assumed that the amount of activated liquid also affects the intensity of dpH response.

2. 'Information copies' of drugs. In these tests we used the technology IC Medicals [21]. Downloading and recording the information on a CD was done according to the instructions listed on the site. Both CDs (non-activated and activated) were placed under the thermostats, as shown in Figure 7(c). Aspirin and gibberellic acid were (randomly) selected as such an 'information drugs'. Some examples of obtained data are shown in Figure 12. Similarly to the case of activated water, the activated and non-activated CD-R discs are physically identical objects, which nevertheless produce a significantly non-zero dynamics of dpH . The figure 12(c) shows an example of organic liquid used for pH measurements, which demonstrates the deviation of dpH more than $25000 \mu V$ for 12 hours. Obviously, that organic solvents are more preferred for the measurements, but they are also sensitive to the other (including non-local) factors, see for example [5], [19]. Control measurements with two empty disks were performed again after a few experimental measurements. In this way it was found that under certain conditions, as well as some time after activation, the activation effect of CD disc disappears, as shown in Figure 15.

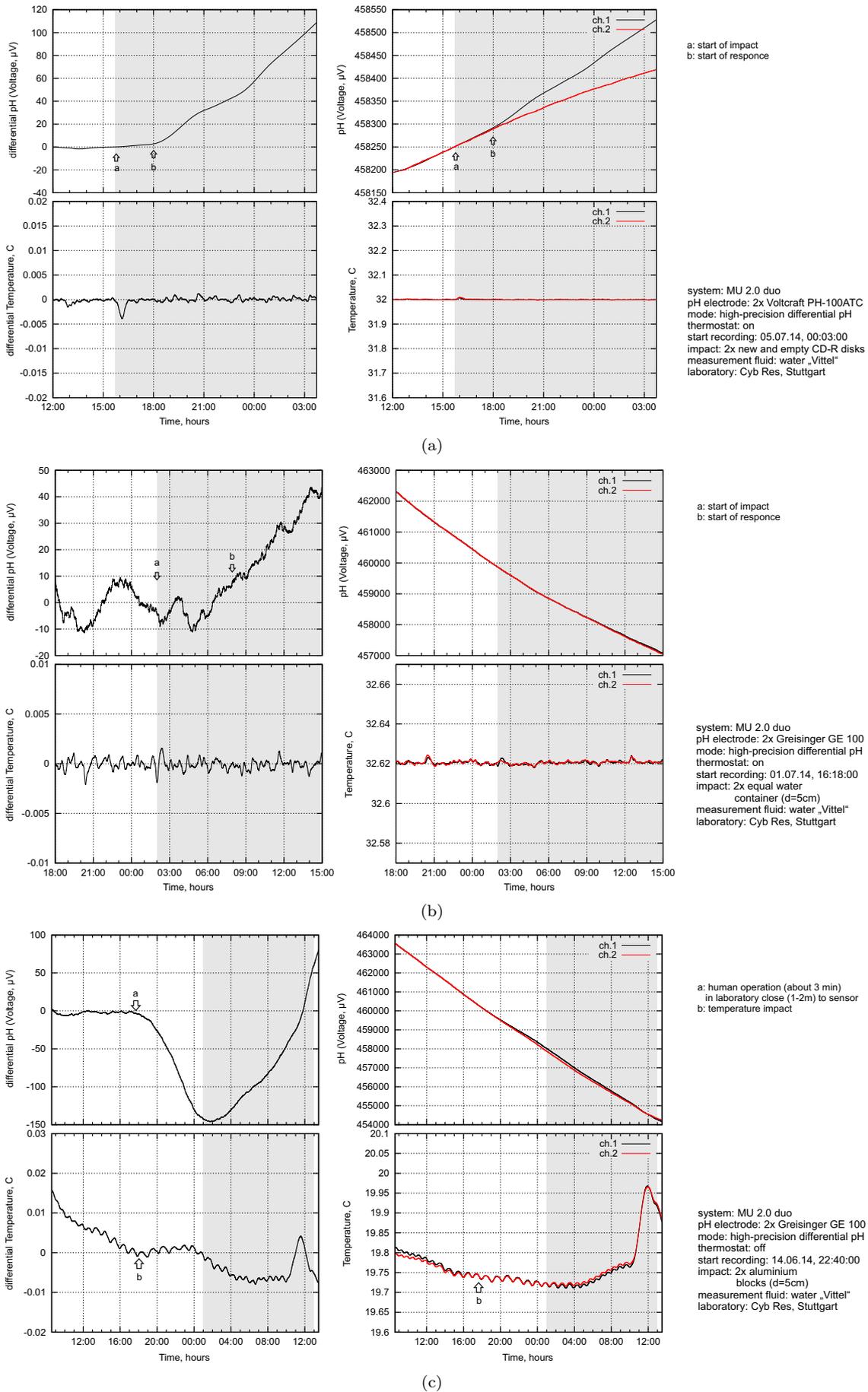


Fig. 8. Control measurement of two identical: (a) CD disks; (b) containers with water; (c) aluminium blocks.

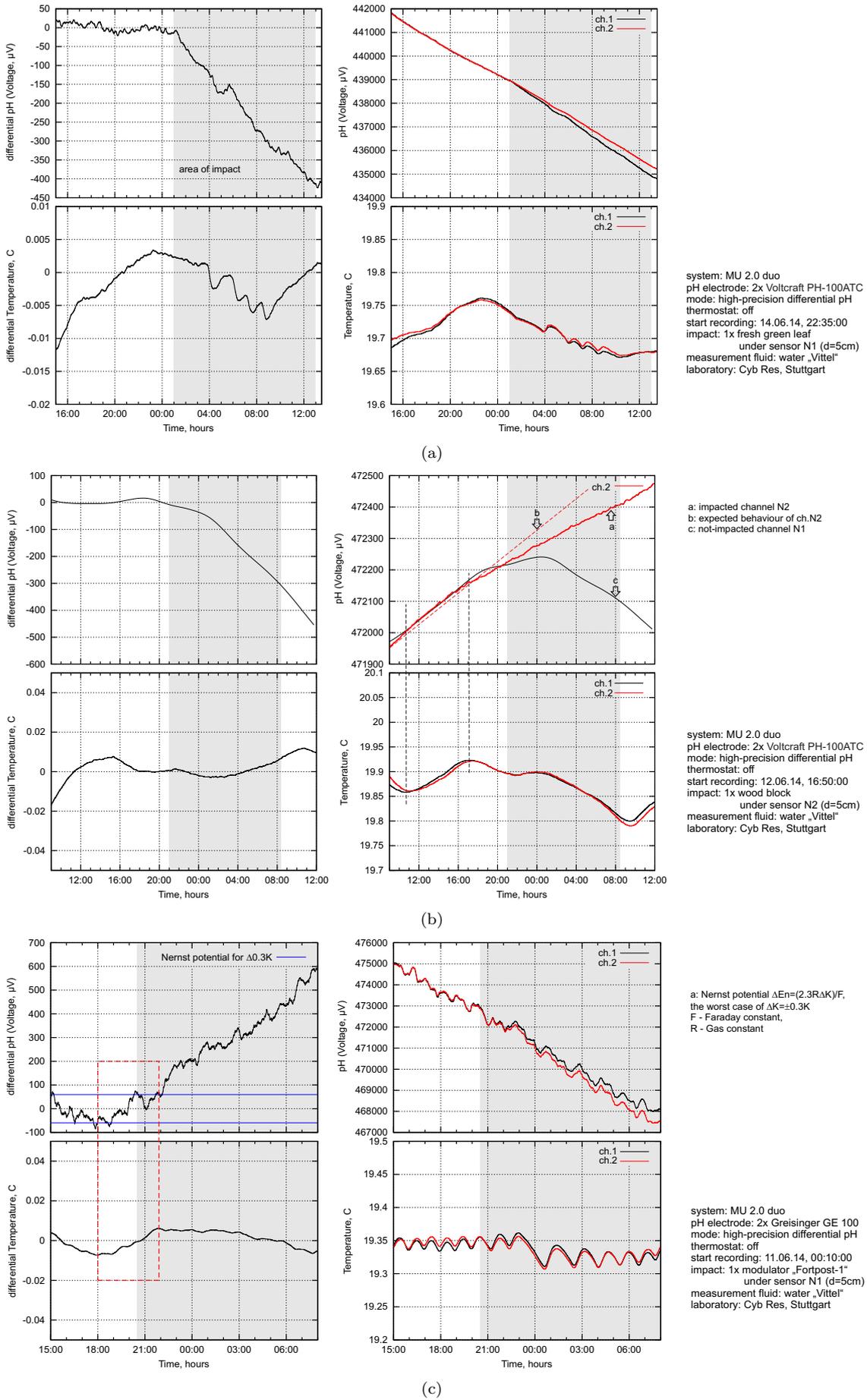


Fig. 9. Control measurement of one object: (a) fresh green leaf; (b) wood block; (c) modulator 'Fortpost-1'.

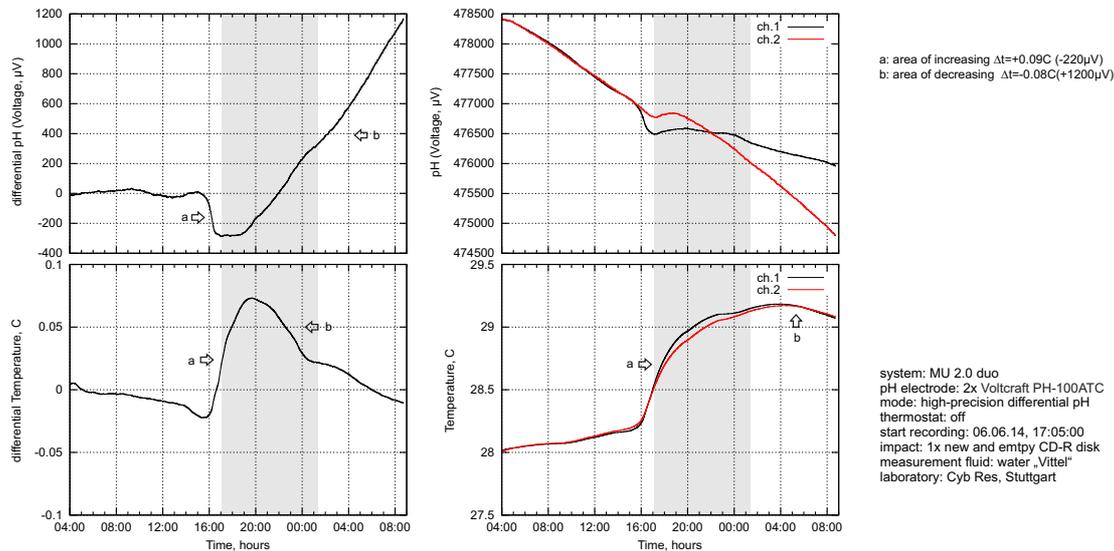


Fig. 10. Control measurement of one CD-R disk.

D. Using of automated differential calibration

Since the program, which automatically evaluates the expression (12), was not ready until the end of this work (when all other measurements have been already done), additional measurements were carried out to demonstrate the automated differential calibration. The user must enter only the time when the measurement and exposure were started, all other conversions are performed automatically.

As a source of impact we used the LED generator with penicillin matrix, the water was activated in 200 ml containers with a closing iron lid, all other experimental parameters were as described above. The parameter Δy in expressions (10), (11) was set to $500 \mu V$ in 12 hours, which corresponds to the electrode drift $\sim 0.001 pH$ for 12 hours. The figure 13 shows examples of the response to these three exposure. Here three dpH device operated in parallel with three different sets of electrodes: no name of Taiwan production, GE100 and HI1131B. Six water containers are used: 3 non-activated and 3 activated as described above. In all three cases we note a similar response at $500 \mu V$ 24 per hour (about $1 mV$ for 36 hours). If case of 'Taiwan' no-name electrodes we observed a noised signal from one of the electrodes (which was also observed in several other experiments.) This suggests a further using of only brand-name laboratory electrodes.

V. SOME CONCLUSIONS

As pointed out by the analysis of literature, the acid-base indicator of different fluids is sensitive at a minimal level to the 'high-penetrating' emission. Considering critically the potentiometric measurement system, it should be noted that a pH electrode has several elements, such as a membrane, the reference liquid or gel, the metal wires (electric double layers polarized in a neighborhood of conductors). It is difficult to say which of these elements act as a sensor at low potentials. The electric double layers in deeply polarized electrodes can represent such

a sensor element [6], [8], [16]. However, in the case of large changes of potentials, such as in organic liquids, we can confidently enough correlate the exposure by 'high-penetrating emission' and variations of acid-base indicator. Here we can confirm the measurements and conclusions stated in the literature.

1. The shown measurements were not intended to obtain statistically significant results – this is a task for future works. The primary aim was to characterize the response of the device to various influences. On the basis of approximately 40 performed measurements we can distinguish three groups of responses. The first group includes background measurements, in which the variation of dpH lies within $\pm 10 \mu V$ (with thermostabilization). The second group includes the use of identical objects with varying dpH about $\pm 100 \mu V$. In the third group has all measurements that use different objects. In this case, a response between $\pm 500 \mu V$ and $\pm 25000 \mu V$ for 12 hours was observed. Thus, we can speak about qualitatively different types of instrument's response in all three cases.

2. Figure 14 shows a comparison between differential dynamics in three cases: (a) non-activated and (b, c) differently activated liquid. There is almost a 10- and 20-fold difference of dpH in measuring liquid between the cases of 'no impact' and 'there is an impact' on the test liquid. The impact starts immediately after installation under the thermostat, changes occur gradually over 12 hours. Full decay time is not yet known. We emphasize again that both measurements were made under the same conditions; the only difference consists in the activation mode of one of the test fluids by LED generator. Since chemical, thermal and electromagnetic transmission factors were eliminated, we are now faced with some new factors that influence the activity of hydronium ions H_3O^+ in measuring liquids. It should also be noted that the activated test fluid is in turn a source of exposure for measuring liquid – this is highly

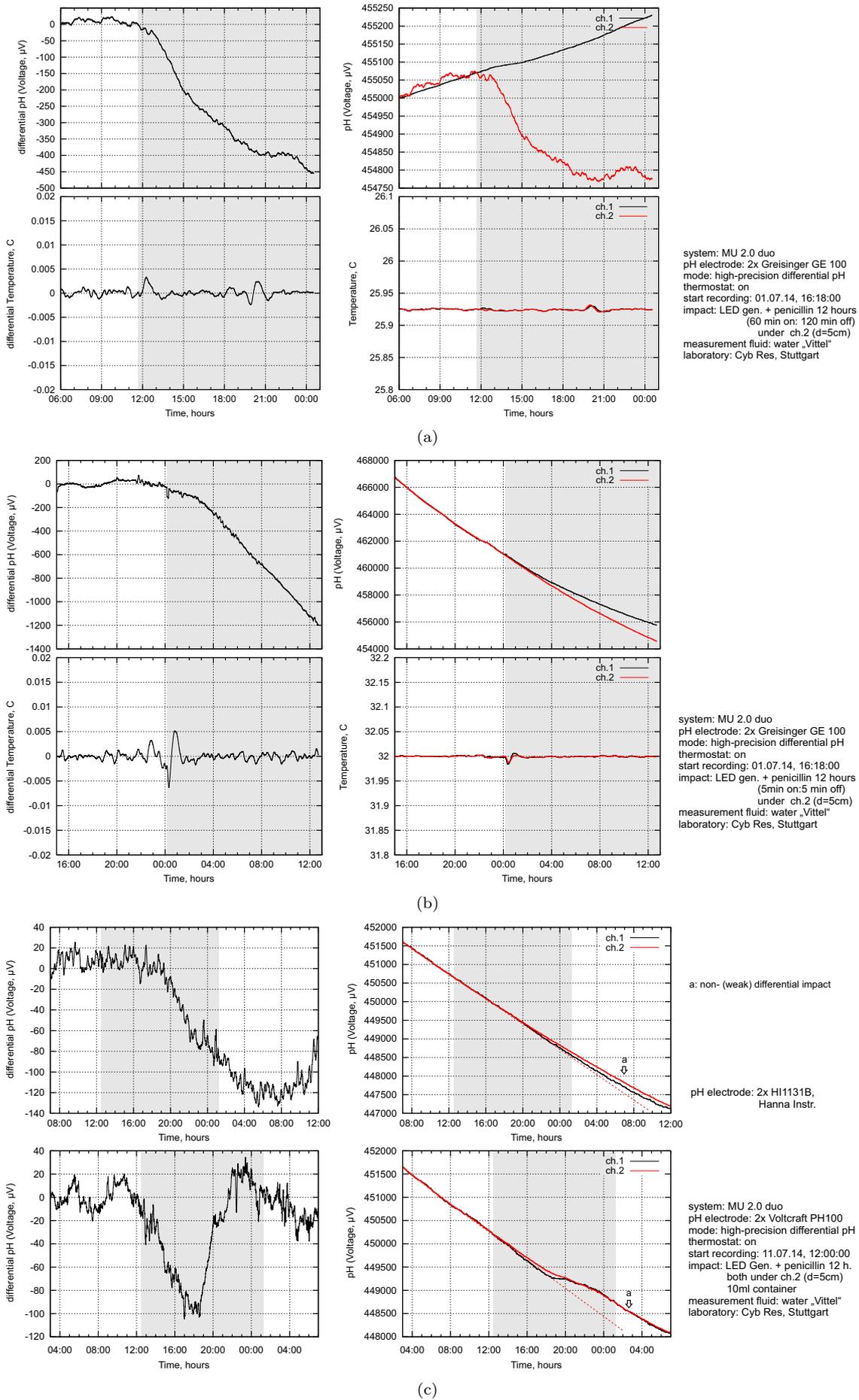


Fig. 11. Examples of a device’s response on test fluid activated by the LED generator.

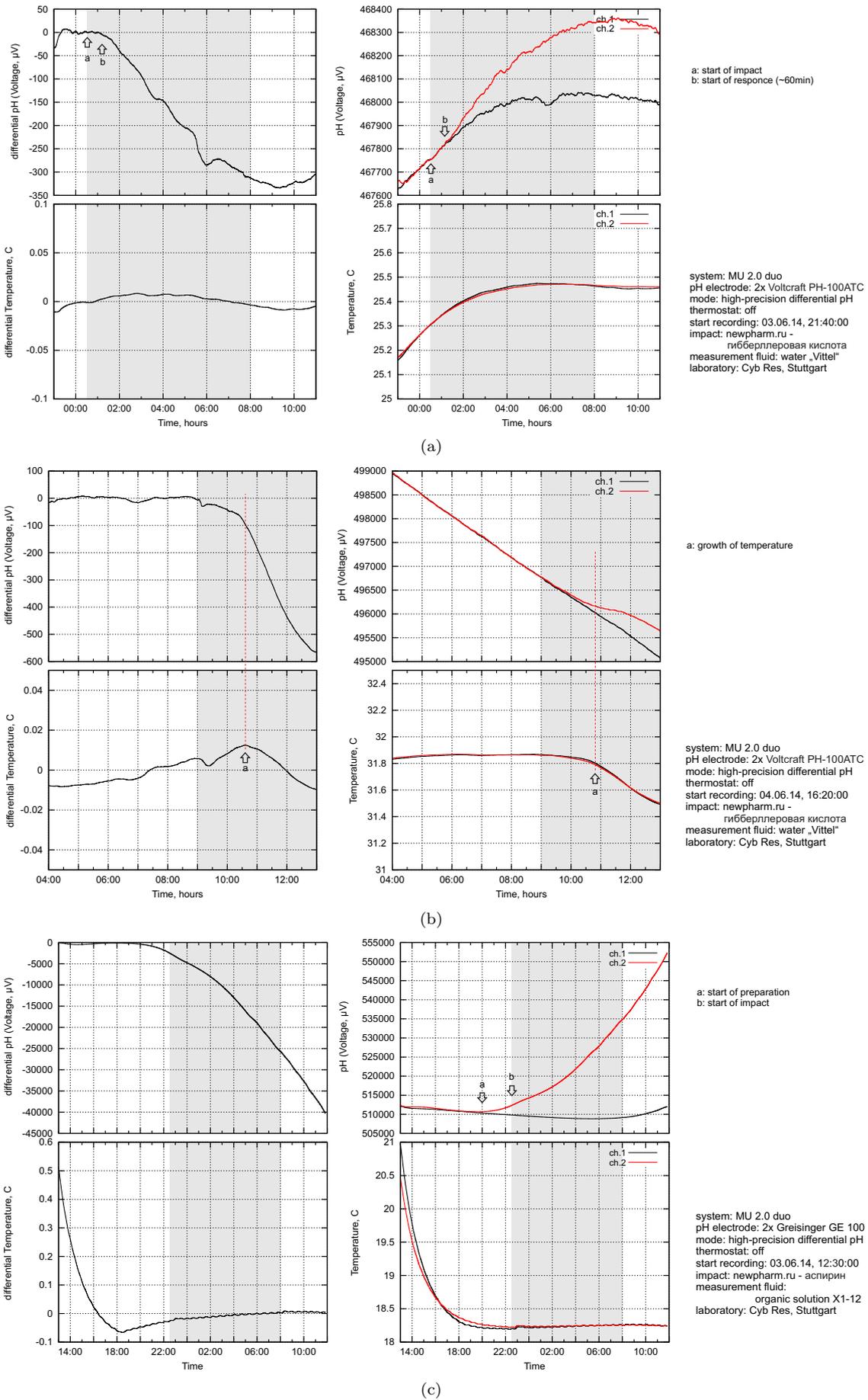
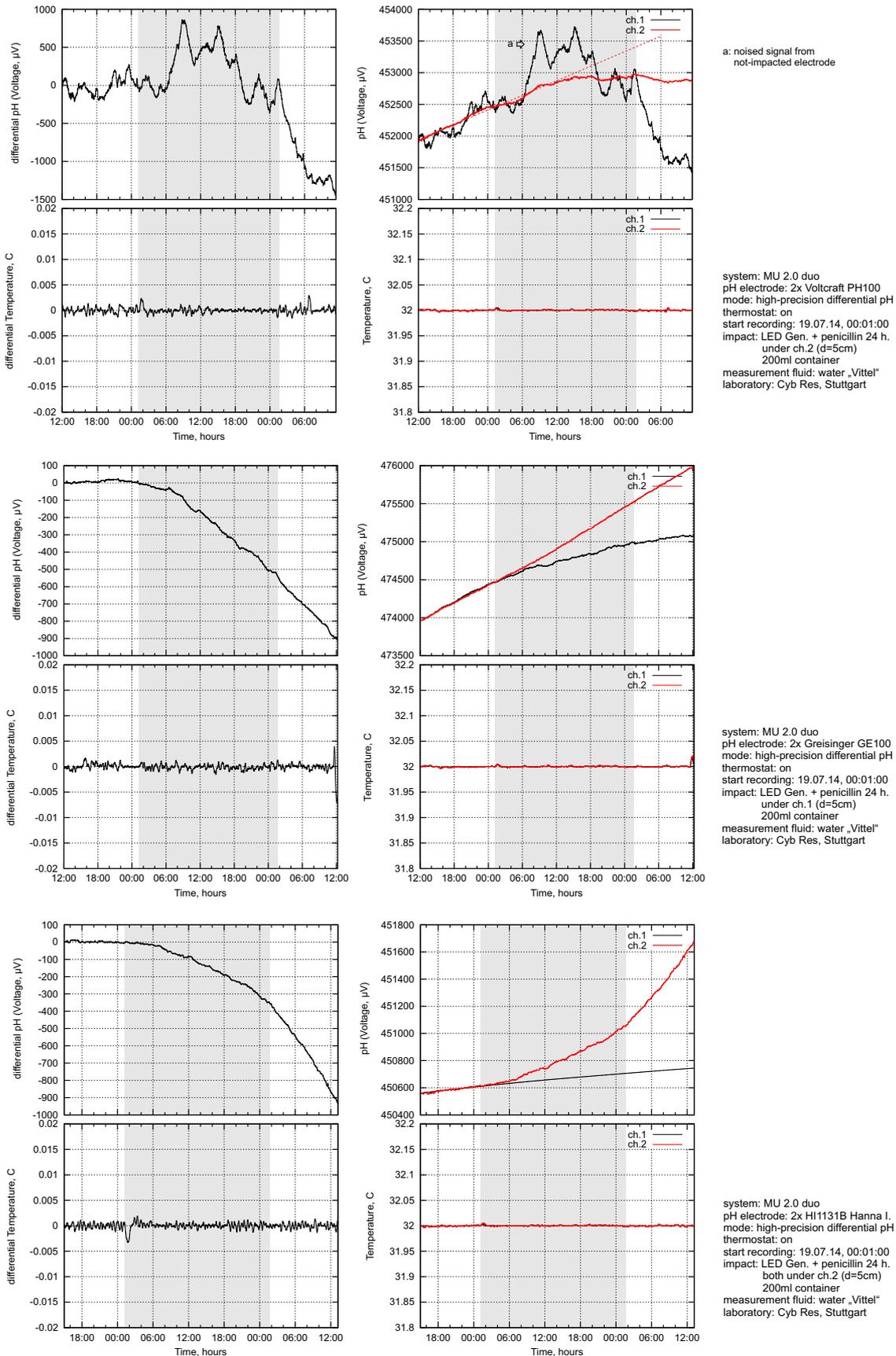


Fig. 12. Examples of a device's response on two physically identical CD-R disks, where one of them is activated in 'informational way'.



(a)

Fig. 13. Example of responses of three working in parallel dpH devices with three different sets of electrodes and with water containers of 200ml by performing automated balancing approach.

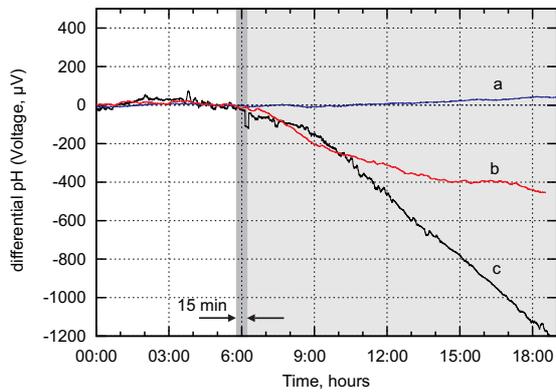


Fig. 14. Comparison of differential dynamics in cases of: (a) identical non-activated fluids; (b) one of fluids is exposed by the LED generator in the mode 60 min. emission: 120 min pause (total exposure time 12 hours); one of fluids is exposed by the LED generator in the mode of 5 min. emission: 5 min. pause (total exposure time 12 hours).

nontrivial for non-ionizing emissions and requires further discussion.

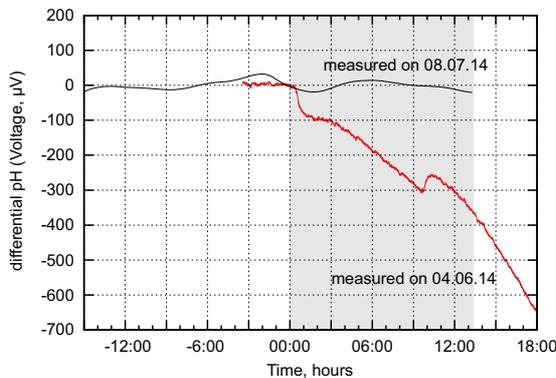


Fig. 15. Comparisons of differential dynamics of activated CD-R disk with info-aspirin (disk was activated at 04.06.14): (a) original measurement on 04.06.14; (b) repeated measurement of the same disk on 08.07.14 (33 days after).

3. Figure 15 shows the measurement of the same CD-R disc, activated on 04.06.14 by 'info-aspirin' (the second CD-R disc was not activated). This measurement was repeated 33 days later on 07.08.14 with the same set of disks. If the initial measurement recorded a deviation of dpH at $400 \mu V$ during 12 hours, then a month later, the effect remained on the level of two non-activated disks. In other words, the activation effect was lost.

4. In the experiments we have encountered the spatial effect by impacting the pH sensors. Two working LED generators were located in the opposite corner of the laboratory at the distance about 2-3 meters away from measuring systems. Both generators used in the PID module, see [19], which – as shown by previous experiments with biological sensors – significantly reduced the external emission from the generators. However, during the dpH measurements it was observed that sensors are impacted by generators, whereupon they were removed from the

laboratory. The Figure 16 shows the response of sensors at the time when generators are removed from the laboratory. It should be noted that the first channel was closer to the generators (distance between channel was about 110 cm). We observed the reaction of both channels, but the first channel showed a larger deviation. Thus, we recorded a spatial difference in the intensity of emission.

5. Almost at the end of this series of experiments, we recorded one interesting effect, as shown in Figure 17. Three devices simultaneously recorded data without any impact during a few days. One of those days was occupied by the final match between football teams of Germany and Argentina on 13.07.14 (where Germany became the champion of world cup 2014 in Brazil).

Emotional atmosphere was very tense before and during the game. We note almost flat dpH and pH dynamics for 10-12 hours prior to 12:00 13.07.14. However, about 12:00 the deviation of pH values between two electrodes began to grow, which lasted up to 2:00-3:00 of the night (match started at 21:00 of local time and lasted until about 0:15). Notably, all three devices showed a deviation at 12:00 almost at the same time. We have no explanation for why the dynamics started to grow 9 hours before the match. It seems there are some analogy with experiments in RNG network, which recorded deviations in behavior of random number generators also some time before global emotional events [32], [33], [34].

To sum up these cases, the device clearly responds to:

- the presence or absence of 'high-penetrating emission'. This applies both to primary (generators) and to secondary (activated objects) sources of emission. For example, activated objects (CD-R discs) lose the activation effect after some time, as shown in Figure 15.
- radiation intensity. It has been demonstrated that the different modes of the LED generator produce the activation of different intensity in the test liquids, as shown in Figure 14.
- spatial non-uniformity of the radiation caused by artificial (generators) and natural (geobiological anomalies) sources of emission. The deviations in dpH dynamics indicates differences in spatial intensity of emission, see Figure 16.
- some emission from the non-activated objects (if only one object is used for dpH measurement). At present, we cannot say anything about the nature and properties of this emission. Comparing graphs in Figures 9 and 10 indicates that different non-activated objects produce also a different response dpH . This could point to different intensity of this emission.
- local and nonlocal activities of operators and global emotional atmosphere (see Figure 17). As shown in Figures 8(c) and 12(c), the deviation of dpH starts at the moments of preparation for the experiment, and also in the presence of operator in the laboratory in excited emotional states.

In the process of clarification and additional experiments are

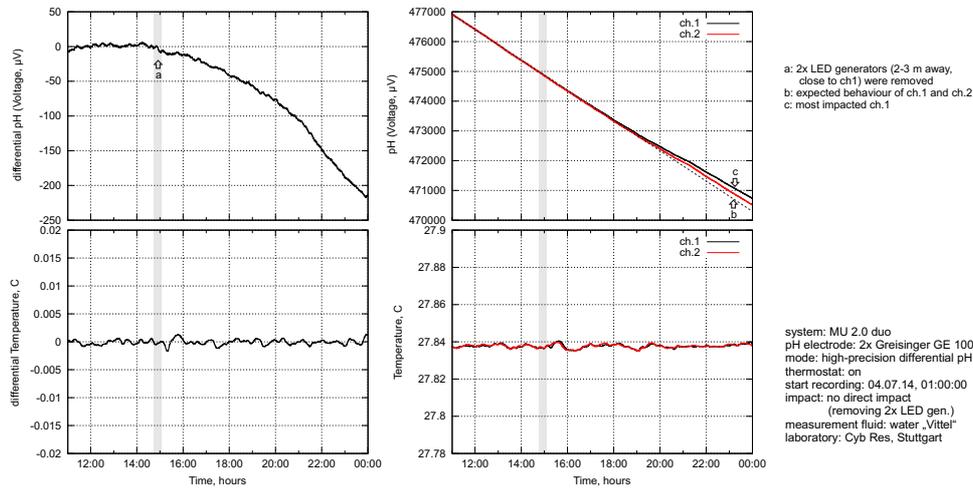


Fig. 16. Reaction of dpH on removing two working LED generators from the laboratory. Gray bar shows the removal time. No essential temperature effects was registered during this time.

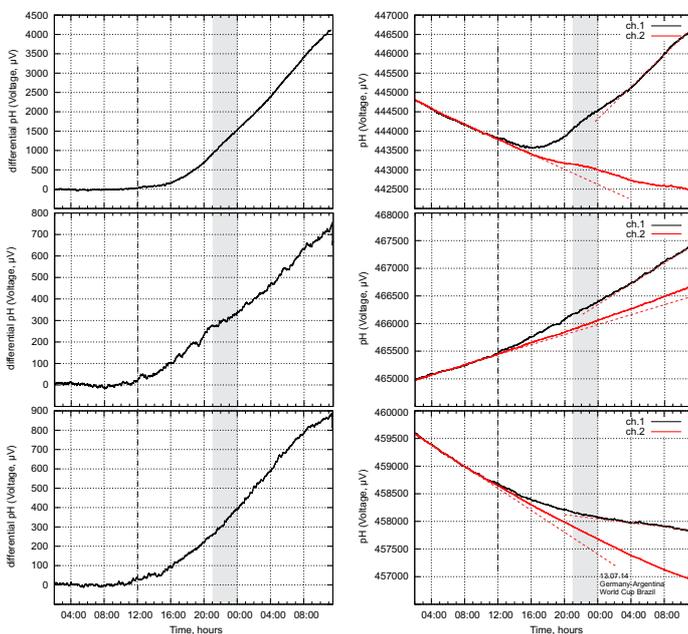


Fig. 17. Data from three in parallel recording dpH devices during the final soccer game between Germany and Argentina on 13.07.14.

- repeatability for different electrodes, different times of day and different spatial positions/orientations of the device. Currently we use three different sets of electrodes: no-name electrodes made in Taiwan (replacement electrodes to the pH -meter Voltcraft PH100-ATC, made in China), electrodes GE100, produced by Greisinger Electronic (GHM Messtechnik GmbH, Germany) and HI1131B, produced by Hanna Instruments, USA, as shown in Figure 18. Preliminary experiments show the dependence of results on the used electrodes (especially for no-name electrodes), and on environmental conditions.
- thermal instability due to the different environmental sources. Only dual temperature control, such as a

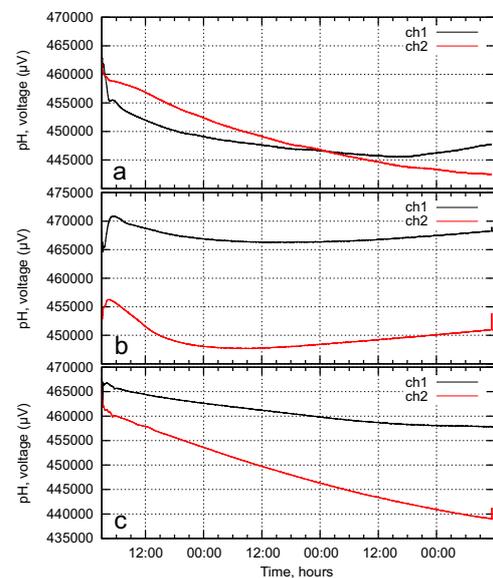


Fig. 18. Dynamics of different uncalibrated pH electrodes during 72 hours: (a) no-name electrodes made in Taiwan, (b) GE100, produced by GHM Messtechnik GmbH, Germany, (c) HI1131B, produced by Hanna Instruments, USA.

thermostatic oven and the internal thermostat, are able to provide excellent thermal insulation. Since experiments require changing the system (i.e. insertion of activated objects), we restrict ourselves to the internal thermostat that ensures the differential zero drift at $\pm 5\mu V$ ($\sim \pm 0.0001pH$) and the differential resolution of $\pm 0.5\mu V$ ($\sim \pm 0.00001pH$). We are also investigating the impact of active thermostat on accurate measurement.

- combination of pH and dpH measurements. In case of strong or 'global' effects (such as global emotional events) both channels demonstrate a reaction. It needs to develop a procedure that combines both pH ,

and dpH measurement with automated differential balancing.

These and other points represent the issues for further works with the device and the dpH measurements.

VI. ACKNOWLEDGMENT

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