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A Stand-Alone Portable Potentiostat With Parallel Channels for Smart Electrochemical Analyses

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Abstract-In recent years portable potentiostats have 1 increased in popularity allowing to perform electrochemical 2 analyses outside the laboratories, moving them to home or 3 point of care (PoC) environments. In this context, the idea of 4 performing multiple acquisitions at the same time is particularly 5 attractive to deal with replicates or with simultaneous multiple 6 quantitative assessments of different analytes, shortening the time required for the analyses and/or increasing data reliability. 8 Multiple parallel channels on a compact and wireless device can 9 maximize the overall system usability. In this article, a multichan-10 nel Wi-Fi-based portable potentiostat is described. The device 11 12 features four independent channels, which can be conditioned with different voltages. The device was designed to minimize the 13 component count and the power consumption, obtaining 23.5 mW 14 per channel. The system was electrically characterized, obtaining 15 an excellent linearity ($R^2 = 0.9999$), a maximum channel-to-16 channel mismatch of 1% when the maximum current range is 17 selected, and a negligible crosstalk among the channels. Moreover, 18 the multichannel potentiostat was tested in the real case scenario 19 of a semiquantitative evaluation of the anti-tissue transglutami-20 nase target antibodies in celiac disease. Two replicates of IgG and 21 IgA were simultaneously acquired, showing a good capability of 22 separating the positive and the negative samples, with a reduction 23 of the acquisition time of 76% with respect to a single channel 24 solution. 25

Index Terms—Chemical sensors, electrochemical devices, mul tichannel, potentiostat, Wi-Fi sensors, wireless sensors.

I. INTRODUCTION

HERE are several contexts in which portable and reliable 29 devices for electrochemical analysis are exploited to pro-30 vide easy-to-use, rapid, highly selective, and quantitative infor-31 mation on target analytes. Possible areas of application are 32 the detection of biological contaminants in food [1], [2], [3], 33 environmental monitoring [4], [5], [6], and clinical diagnostics 34 [7], [8], [9], [10], [11]. Many of them are based on the use 35 of a potentiostat, i.e., an electronic hardware elaborating the 36 electrical signal generated from an electrochemical reaction. 37

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With reference to this, the main challenge tackled by the researchers is how to implement a compact portable device with cheap components and reduced power consumption without compromising the quality of the tests. It is worth noting that the overall portability of the device strongly depends also on the architecture of the system. In particular, for the development of a completely stand-alone instrument, it is preferable to avoid the use of additional devices for data elaboration and storage. Recently, several portable solutions have been commercialized [12], [13], [14]; however, for their operation, they rely on supplementary devices such as personal computers (PCs) or tablets/smartphones connected via USB and/or Bluetooth. Also, some interesting solutions are available in the literature [4], [15], [16], [17] with some advanced features like the connection to a cloud service, but all require the connection to an external device, limiting the portability and the overall usability.

Moreover, these solutions, as most of the available portable potentiostats, are designed for single analyte detection and are equipped with a single analog front end (AFE) to read and control the electrochemical sensor [4], [15], [16], [17], [18]. Many of these electrochemical acquisitions are timeexpensive, with a single experiment easily lasting several minutes. Moreover, replicates are needed to increase the result precision to obtain statistically relevant results. Furthermore, sensor redundancy allows the implementation of advanced fault-tolerant algorithms and data-averaging techniques that can improve system robustness, efficiency, and accuracy [19]. Bioanalytical, pharmaceutical, and clinical applications require simultaneous electrochemical detection that cannot be obtained using a single-channel instrument. For example, parallel measurements of multiple cancer markers are useful for increasing the efficiency of cancer diagnostics and therapy monitoring [20], as well as for considering possible correlations between compounds [9]. Therefore, a potentiostat with multiple acquisition channels would significantly increase the efficiency of the experiments, implementing their parallel execution.

In this article, a multi-channel portable potentiostat with three electrodes for each channel suitable for both quantitative and semiquantitative electrochemical analysis, usable in a large variety of out-of-lab applications, is presented. The device features four completely independent and parallel channels, capable of simultaneously managing, with different voltages, four commercial screen-printed electrodes, each of which

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composes a three-electrode electrochemical cell. Thanks to 83 six-channel analog-to-digital converter (ADC), the current а 84 measurement is carried out at the same time for all the 85 channels. The device can be battery-powered and connected, through a Wi-Fi protocol, to a cloud analytics for data storage 87 and elaboration. This allows a simple connection to wide-88 spread Wi-Fi networks for sharing data with different users 89 without the need for external devices. Moreover, the cloud 90 service can be interfaced with a web application for the remote 91 control of the device. The aim of this article is the description 92 and the characterization of the hardware device. A detailed 93 description of a possible web application can be found in [18]. 94 The proposed four-channel solution features a scalable 95 architecture that can be easily expanded to meet user needs.

As a case study, an electrochemical immunosensor for 97 the detection of anti-tissue transglutaminase antibodies 98 (anti-tTG) was considered [21]. These antibodies represent 99 reliable biomarkers for the diagnosis of celiac disease, for 100 which both the IgA and IgM isotype of anti-tTG must be 101 quantified. Indeed, it is common for patients affected by celiac 102 disease to display a selective IgA deficiency which, in the 103 absence of IgG quantification, would lead to a false negative 104 outcome of the diagnosis. Therefore, the use of a multi-channel 105 acquisition device is of paramount importance to allow both 106 the simultaneous determination of the IgA and IgG isotypes 107 of anti-tTG antibodies and the acquisition of at least two 108 replicates during a single measurement cycle. 109

This article is organized as follows. In Section II, the related 110 works are presented; in Section III, the architecture of the 111 system designed is described; in Section IV, the experiments 112 performed to characterize the device are reported and dis-113 cussed; and in Section V, conclusions are drawn. 114

II. RELATED WORKS

The key task of a potentiostat device is to measure the 116 current flowing into an electrochemical cell due to a chemical 117 reaction related to the target analyte. The electrochemical 118 sensing systems require two [22] or three electrodes [7]. 119 A three-electrode electrochemical cell includes a working 120 (WE), a reference (RE), and a counter electrode (CE), while 121 in the two-electrode cell, CE and RE are unified. It is worth 122 123 notingd that, regardless of the number of electrodes in the cell, in order to carry out different types of analyses simultaneously 124 in a multichannel system, it is important to keep independent 125 the electrodes of the different cells under test. In this way, 126 it will be possible to condition the different cells with different 127 voltages according to the analysis to be carried out. 128

In [23], a multichannel potentiostat is presented. Thanks 129 to four ADCs with single-ended inputs, the device is able to 130 read four independent two-electrode cells simultaneously. The 131 core of the system is a Raspberry PI Computer. Through I2C 132 communication, up to eight potentiostats can be connected. 133 It features a 12-bit DAC for generating the control voltages. 134 Moreover, traditional potentiostats rely on a transimpedance 135 amplifier (TIA) for current measurements. In this case, the 136 authors exploited a shunt resistor, lowering the overall accu-137 racy of the device. The maximum current range is $\pm 1.5 \ \mu$ A. 138

It is worth noting that, despite the high number of channels 139 available in this solution considering the eight boards stacked 140 together, the reading of the channels is not simultaneous, but 141 it is serialized through the communication protocol chosen 142 (e.g., I2C). Moreover, notwithstanding the two-electrode sys-143 tem results in a simplified circuitry [22], the three-electrode 144 solution is more advisable since the current deriving from 145 the chemical reaction flows through the CE instead of the 146 reference one, avoiding the change in the potential of the WE 147 [7]. For this reason, the three-electrode solution is the most 148 widely adopted. 149

In [24] and [25], a compact architecture with 128 chan-150 nels is reported, where, to reduce the device dimensions, 151 the single potentiostat channel contains only the essential 152 components while the rest of the hardware is time-shared 153 across channels and driven by a single μ C. To increase the 154 number of channels, a time division multiplexing technique is 155 exploited. The measurement results are sent to a PC through 156 a UART port, limiting the portability of the device. Moreover, 157 the CE and RE are shared between the parallel channels 158 limiting the possibility of conditioning the electrochemical 159 cells with different voltages. The same limitation applies to 160 the 8-channel potentiostat named "octopoti" presented in [24]. 161 This device cannot be considered a portable solution, as it 162 requires a suitable external data acquisition system to read the 163 outputs. Similarly, in [26], a 16-channel potentiostat requiring 164 a multifunction PC with a data acquisition card to be powered 165 and controlled is presented. 166

In [27], a six-channel wireless potentiostat is described. 167 In this case, the solution turns out to be quite portable even if 168 the choice of a low-range communication protocol such as 169 Bluetooth does not eliminate the need to have an external 170 device (e.g., PC, tablet, or smartphone) nearby, which acts 171 as a gateway for the acquired data. Moreover, the data elabo-172 ration and visualization require the LabVIEW data acquisition 173 system running on an external device. 174

In [28], a three semi-parallel channels solution is reported. 175 It requires a USB or Bluetooth connection to operate and 176 relies on a smartphone or desktop APP for data processing and 177 visualization. It features a 12-bit DAC and an ADC embedded 178 into the ESP32 Microcontroller. The ADC is in common for 179 the three channels. For this reason, although each of the three 180 channels has dedicated electronics, the data sampling process 181 is performed sequentially, alternating the reading of each ADC 182 pin. 183

Also, some commercial solutions are available. MultiPalm-184 Sens4 and MultiEmStat4 from PalmSens [12] are benchtop 185 instruments with up to 10 and 12 channels, respectively. The 186 portability of these devices is very low, as they commu-187 nicate over a USB cable and are powered with an exter-188 nal 12V ac/dc adapter or the same USB port. A more 189 portable device is the wireless dual-channel potentiostat 190 Sensit-BT [12]. Another commercial solution is the STAT8000 191 from Metrohm [15], a portable eight-channel potentiostat. 192 Both are battery-powered and feature a Bluetooth inter-193 face. The STAT8000 requires a PC for data elaboration and 194 visualization. 195



Fig. 1. Schematic of the four-channel potentiostat.

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III. SYSTEM DESIGN

The design of the proposed four-channel potentiostat was 197 optimized for power consumption and component count. 198 To this aim, devices available in the dual or quad con-199 figuration were preferred to maintain the same number of 200 components and the same board size of the single-channel 201 implementation [29]. Dedicated ADCs were introduced to 202 improve the quantization error with respect to the A/D con-203 version channel available in the μC and to implement the 204 simultaneous sampling over the four channels. The analog 205 components were selected for the minimum supply current 206 and with dynamic performance (i.e., bandwidth and conversion 207 speed) compatible with the target configuration (i.e., battery 208 powered) and the frequency of the voltage signal and the 209 output current [29], [30]. 210

The schematic of the potentiostat is shown in Fig. 1. The 211 designed AFE is interfaced with a CC3200 μ C [31], a system 212 on a chip (SoC) embedding an IEEE 802.11 compliant Wi-Fi 213 radio [31]. This allows the wireless link capability to be added 214 to the potentiostat device without increasing the component 215 count. Acquired data can be processed on board and then sent 216 to a cloud service for storage and visualization. 217

For the AFE, the single-ended potentiostat (SEP) archi-218 tecture, with only two operational amplifiers (opamps) per 219 channel [29], [32], was selected for the cell control and 220 readout, since it exhibits the minimum component count [18], 221 [32]. The former opamp, i.e., A1, is used to control the 222 CE voltage and set the RE voltage, and the latter, i.e., A2, 223 to implement the transconductance amplifier (TIA) for sensing 224 the cell output current. The cell bias voltage, i.e., $V_{\text{bias}} \equiv$ 225 V_{WE} - V_{RE} (the potentials at the WE and RE pin, respectively), 226 is equal to the difference of the voltage at the positive input 227 pin of A1 and A2, i.e., $V_{\text{bias}} \approx V_Y - V_X$, provided that the 228 opamps operate in the linear region. Two quad-DAC AD5696 229 independently generate for each channel the bias voltage of the 230 TIA, i.e., V_Y , and the voltage signal to be forced at the RE pin, 231 V_X . If the differential pulse voltammetry (DPV) technique is 232 used to assess the concentration of the chemical species, V_X 233 is a pulsed stair-step signal with constant rising and falling 234



Fig. 2. TIA schematic with feedback network involving the chemical cell. The chemical cell has been modeled with its equivalent electrical circuit (dummy cell)

steps [29], [30]. The DACs are controlled by the μ C through a standard I2C bus.

The AD8608 quad opamp exhibits an input current within tens of picoampere, not to perturb the chemical reaction at the RE pin, and an adequate output current capability to guarantee a short pre-conditioning time of the chemical cell [33].

The TIA opamp A2 is a critical device since it sets the 241 equivalent input noise current of the readout channel. Further-242 more, its stability margin is heavily impacted by the RE-WE 243 capacitance (C_{RW}) in the lumped-circuit electrical model of 244 the chemical cell, Fig. 2 [34].

Indeed, this capacitance may be as high as 10 μ F, depending 246 on the cell area implementation and the chemical species [35], 247 leading to potential stability issues for the TIA. The schematic 248 circuit for the evaluation of the stability margin and the con-249 straints on the unity-gain bandwidth (GBW) of the involved 250 opamp is shown in Fig. 2. 251

The equivalent impedance from CE pin to ground (Z_{CE}) 252 usually has a negligible effect on the TIA stability margin, 253 and it is neglected in the following analysis. The feedback 254 network, involving C_T , R_T , and the potentiostat equivalent 255 circuit, introduces a pole and a zero in the loop gain transfer 256 function, T(f). The corresponding frequencies f_{pr} and f_{zr} 257 are approximated by the following equations: 258

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$$f_{pr} \cong \frac{1}{2\pi \left(C_{\text{RW}} R_H\right)} \tag{1} 25$$

$$f_{pz} \cong \frac{1}{2\pi \left(C_T R_T\right)} \tag{2}$$

where R_H is the resistance value of the parallel combination 261 of $R_{\rm RW}$ and R_T . In the proposed implementation, the TIA 262 feedback resistance is set within the range from 10 k Ω 263 to 1 M Ω . 264

The gain programmability is implemented with a pair of 265 four-way solid-state MUXs that allow the selection of one 266 feedback resistance over eight available values [18]. The 267 MUXs are driven by the μC through the general-purpose 268 input-output (GPIO) ports to continuously adapt the TIA gain 269 during the measurement. This background gain calibration 270 aims at maximizing the signal swing at the ADC input, always 271 keeping the TIA opamp in the high-gain region and avoiding 272 the ADC saturation. 273

The minimum GBW of the TIA opamp is calcu-274 lated from (1) and (2) to obtain a phase margin higher 275

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Fig. 3. Voltage waveforms at the TIA output and ADC input (zero-mean signal case). (a) Without BLS. Case 1: $V_Y < V_{M-TIA}$; Case 2: $V_Y < V_{M-TIA}$; (b) With BLS between ADC and DAC. Cases 1 and 2: $V_Y < V_{M-TIA}$; cases 3 and 4: $V_Y < V_{M-TIA}$. In cases 1 and 3 the reading range is TIA limited, and in 2 and 4 is ADC limited.

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$$\text{GBW}_{\text{A2}} > \frac{C_{\text{RW}}R_H}{2\pi \left(C_T R_T\right)^2}.$$
(3)

For the present design, the LTC6082 (quad opamp) was used. It exhibits a typical GBW of 3.5 MHz with a current consumption of 330 μ A and can be compensated over the range of feedback resistors.

In the design of the readout channel of the current flowing into the WE terminal (I_W), the signal swing at the TIA output should be carefully considered. Indeed, the reading range of the potentiostat cannot exceed the boundaries set by the output range of opamp A2, at a given value of feedback resistance R_T

$$V_{\text{OMIN}} - V_Y \le I_W \cdot R_T \le V_{\text{OMAX}} - V_Y \tag{4}$$

where V_{OMAX} and V_{OMIN} are the maximum and minimum output voltage of the TIA opamp, corresponding, for the LTC6082, to 3.3 V supply minus 30 and 30 mV, respectively. A further constraint is introduced by the conversion range of the ADC, assumed from 0 to V_{ADC}

$$0 < V_Y + I_W \cdot R_T < V_{\text{ADC}}.$$
 (5)

Restricting the analysis to the case of a zero-mean cur-294 rent signal, the reading range is limited by the lower 295 bound of the TIA-opamp (output) voltage in the case 296 where V_Y is lower than the midpoint of the opamp output 297 range, i.e., $V_{M-TIA} = 0.5 \cdot (V_{OMAX} - V_{OMIN})$, as shown in 298 Fig. 3(a). With V_Y higher than V_{M-TIA} , the reading range is 299 bounded by the ADC conversion range, provided that V_{ADC} is 300 lower than V_{OMAX} . 301

³⁰² A further relevant design aspect is the signal-to-³⁰³ quantization-noise ratio (SQNR) of the readout channel. The ³⁰⁴ maximization of the SQNR requires maximizing the signal ³⁰⁵ swing at the ADC input as well. Still assuming a zero-mean ³⁰⁶ $I_W(t)$ signal, the SQNR is estimated by the following formula:

sor SQNR =
$$10 \cdot \log \left[\frac{3 \cdot 2^{2 \cdot N_B} \cdot (\alpha_W \cdot \Delta I_W \cdot R_T)^2}{V_{ADC}^2} \right]$$
 (6)

where N_B is the nominal resolution of the ADC, ΔI_W is the peak-to-peak swing of the cell current, and α_w is equal to one for a square-wave input current or to $1/\sqrt{3}$ for a triangular waveform.

As shown in Fig. 3(b), the mismatch between V_Y , V_{M-TIA} , and the midpoint of the ADC range prohibits achieving the



Fig. 4. BLS interfacing the TIA to the ADC.

maximum SQNR in (6) with $\Delta I_W \cdot R_T = V_{ADC}$. The problem 314 has been addressed in the proposed design with a dedicated 315 buffer-level shifter (BLS) placed between the TIA and the 316 ADC. As shown in the schematic of Fig. 4, this buffer requires 317 one additional opamp per channel (A3), whereas the opamp 318 A4 is used to buffer a reference voltage (V_{MID}) derived from 319 the supply and shared over all the channels. Furthermore, since 320 the bandwidth of A3 should be compatible with the settling 321 time of the $I_W(t)$ signal, and thus, smaller than the TIA-opamp 322 GBW, the LTC6079 featuring a current consumption of 54 μ A 323 was used. 324

With the resistance settings as in Fig. 4, the buffer output voltage, corresponding to the ADC input signal, is centered at the V_{MID} voltage value and amplified by G

$$V_{\text{IN-ADC}} = G \cdot (V_{O-\text{TIA}} - V_Y) + V_{\text{MID}} = G \cdot I_W \cdot R_T. \quad (7) \quad {}_{328}$$

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The reference voltage V_{MID} must be equal to the midpoint 329 of the ADC range, i.e., 1.25 V in our design. 330

The reading range ΔI_W at the selected TIA gain R_T can be either limited by the TIA or the ADC, depending on the value of *G* and V_Y . If the ADC saturation occurs before the TIA-opamp saturation, the reading range is ADC-limited, as in cases 2 and 4 in Fig. 3(b). Depending on the setting of V_Y with respect to V_{M-TIA} , the condition for ADC-limited range is

$$V_Y > V_{\text{M-TIA}} \rightarrow G \cdot (V_{\text{OMAX}} - V_Y) \ge \frac{V_{\text{ADC}}}{2}$$
 (8) 33

$$V_Y > V_{\text{M-TIA}} \rightarrow G \cdot (V_Y - V_{\text{OMIN}}) \ge \frac{V_{\text{ADC}}}{2}.$$
 (9) 338

At the measurement startup, the μ C will set the TIA gain at the maximum value. During the measurement, the gain will be decreased as soon as the ADC output approaches the upper or the lower saturation condition, corresponding to conditions (8) and (9), respectively. 343

If neither condition (8) nor (9) is fulfilled, the reading range is TIA-limited. In this case, the μ C will decrease the gain during the measurement at the occurrence of the following condition for the ADC output code D_{O-ADC}: 347

$$D_{\text{O}-\text{ADC}} = 2^{N_B - 1} + k_O \cdot \left(2^{N_B} - 1\right) \cdot \frac{G \cdot \Delta V_O}{V_{\text{ADC}}} \qquad (10) \quad \text{346}$$

CURRENT CONS. BREAKDOWN



Fig. 5. Current consumption breakdown of the proposed four channels potentiostat. The consumption of the readout circuits for the WE and RE voltage (WE/RE V-SENS) includes the voltage buffer and the related ADC.

where ΔV_O and k_O depends on the position of the V_Y level 349 in the TIA output range, as in cases 1 and 3 in Fig. 3(b) 350

$$V_Y > V_{M-TIA} \to \Delta V_O = (V_{OMAX} - V_Y), \quad k_O = +1 \quad (11)$$

$$V_Y \le V_{\rm M-TIA} \to \Delta V_O = (V_Y - V_{\rm OMIN}), \quad k_O = -1.$$
 (12)

In the present design, we used the LTC1408 device inte-353 grating six A/D conversion channels. The device was selected 354 for the simultaneous sampling capability, the low power 355 consumption, and the 14-bit nominal resolution, which is 356 suitable for the present potenstiostat [36]. The μ C provides 357 the sampling and the clock signals to the ADC through 358 a three-wire serial interface. The same interface allows the 359 acquisition of the output data stream. It is worth noting that the 360 outputs of the four channels have been connected to the same 361 ADC. Hence, the sampling of the outputs of all the channels 362 happens simultaneously. The results are then transmitted to 363 the microcontroller with a serial interface, but this does not 364 affect the timing of the measurements since the transmission 365 is completed between two subsequent measurements. 366

Additionally, the potentiostat allows the acquisition of the 367 voltages at the WE and RE pins for each cell. These readout 368 channels (not shown in Fig. 1) add a diagnostic capability to 369 the system since the virtual short circuit at the input of A1 370 and A2 opamp can be continuously monitored during the cell 371 current acquisition cycle. Furthermore, they allow measuring 372 the voltage across WE and RE terminals, which is required in 373 the DPV measurement procedure [29], [30]. 374

The current consumption of the AFE (excluding the μ C) 375 is 3.3 mA per channel. A regulated 3.3 V is provided by an 376 onboard low-dropout (LDO) regulator from either the battery 377 or the 5 V USB supply. The consumption breakdown is 378 obtained with circuit simulation (using the SPICE models pro-379 vided by the manufacturers) and from the current consumption 380 values reported in the component data sheets [36]. The results 381 are shown in the graph in Fig. 5. 382

With regard to the digital part of the system, a detailed 383 analysis of the power consumption was discussed in [29]. 384 Here, considering the worst case of continuous operation with 385 the μ C always active, without low-power mode management 386 between two readings, a current of 3.8 mA per channel should 387

TABLE I CURRENT RANGE, RESOLUTION, AND INPUT NOISE

RT	$\pm \Delta I_W/2$	Resolution	I _{IN-N} rms		
(kΩ)	(μΑ)	<u>(nA)</u>	(nA)		
10	60	7.6	1.72		
100	6.0	0.76	0.173		
1000	0.60	0.076	0.017		
TABLE II					
CHANNEL MISMATCH RESULTS					

R _{RW}	EA,max	Es	Ei	r ² min
<u>(Ω)</u>	(mV)	(%)	(%)	
10k	9.2	1	0.5	0.9999
100k	5	0.9	0.4	0.9999
1M	3.1	0.07	0.3	0.9999

be considered. Hence, the total current required for both the analog and digital parts is 7.1 mA.

The reading range, the resolution, and the simulated input-current noise (I_{IN-N}) , root mean square (rms), for the 391 minimum, i.e., 10 mV/mA, medium, i.e., 100 mV/mA, and maximum, i.e., 1 V/mA, TIA gain are reported in Table I.

IV. RESULTS AND DISCUSSION

A. System Characterization

Several tests were carried out to evaluate the performance 396 of the proposed multichannel portable potentiostat. The single-397 channel performance was evaluated to assess both the linearity 398 of the response and the output difference among channels with 399 the same voltage signals and load (i.e., channel mismatch). For 400 that purpose, a dummy cell was connected to the input of each 401 channel. A schematic of the dummy cell is reported in the red 402 box in Fig. 2. 403

Three different values of $R_{\rm RW}$ were taken into account (i.e., 404 10 k Ω , 100 k Ω , and 1 M Ω). This allows considering differ-405 ent input current ranges (i.e., maximum currents of 62 μ A, 406 6.2 μ A, and 620 nA, respectively). The considered ranges are 407 those usually needed in some common applications. For each 408 $R_{\rm RW}$ value, each DAC channel was configured to provide a 409 voltage bias ranging from -0.6 to 0.6 V, and the corresponding 410 Vout, i.e., the digital ADC output expressed in volts, was 411 acquired. For each channel combination, an absolute error 412 ε_A was evaluated, corresponding to the difference between 413 the measured V_{out} of two considered channels. In Fig. 6, the 414 measured absolute error between channels is shown, with an 415 $R_{\rm RW}$ value of 1 M Ω . 416

This analysis was repeated for each possible value of R_{RW} , evaluating the maximum error in the measurement of the output voltage (Table II).

Moreover, a relative error in the slope and the intercept 420 of the fit curve was evaluated as a further parameter of the 421 mismatch between different channels. The relative error in the 422 slope ε_s was defined as 423

$$\varepsilon_s = \frac{m_{\rm max} - m_{\rm min}}{m_{\rm avg}} \tag{13}$$

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Fig. 6. Absolute error, corresponding to the difference between the measured V_{out} of the two considered channels, when the 1 M Ω resistor was selected into the dummy cell.

where $m_{\rm max}$ is the maximum value of the fit curve slope, 425 among the four-channel data, given a dummy cell resistance 426 value, m_{\min} is the minimum value, and m_{avg} is the averaged 427 value of the four-channel slopes. 428

Similarly, the relative error in the intercept ε_i was defined 429 as 430

$$\varepsilon_i = \frac{q_{\max} - q_{\min}}{q_{\text{avg}}} \tag{14}$$

where q_{max} is the maximum value of the fit curve intercept, 432 among the four-channel data, given a dummy cell resistance 433 value, q_{\min} is the minimum value, and q_{avg} is the averaged 434 value of the four-channel intercepts. The overall results are 435 reported in Table II. 436

Finally, the linearity in the V out measurements was evalu-437 ated. An example of such an evaluation is reported in Fig. 7, 438 when an $R_{\rm RW}$ value of 1 M Ω is selected. 439

These reported values show that each channel of the pro-440 posed multichannel potentiostat exhibits excellent linearity 441 442 regardless of the resistance chosen on the dummy cell and then the current sensed by the device. The maximum error 443 due to the channel design difference (e.g., components value 444 and PCB design) is 1%. This is compatible with the tolerance 445 chosen for the gain and feedback resistor. 446

Once the maximum mismatch among the channels was 447 assessed, tests were carried out to quantify the channel-to-448 channel crosstalk. As the channels showed negligible mis-449 match, only channels 1 and 2 were considered for these tests. 450 Two identical dummy cells (Fig. 2) were connected to the 451 channels and configured with a resistance value of 10 k Ω . 452 The onboard DAC was used to generate a sinewave signal with 453 0.3 V amplitude and 0.5 V offset at different frequencies (i.e., 454 1, 5, 10, 25, 50, and 100 Hz). This signal was applied to the 455 V_X pin of channel 1 in Fig. 1 and thus replicated to $V_{\rm RE}$ (due 456 to the virtual short circuit at the A1 input). The channel 1 TIA 457 bias voltage V_Y was set to 0.5 V. The V_X and V_Y inputs of 458



Fig. 7. Linearity in the four-channel measurements when a 1 M Ω resistor is selected in the dummy cell.

TABLE III CHANNEL CROSSTALK RESULTS

Input signal frequency (Hz)	FFT amplitude ratio (x1000)		
1	2.6		
5	4.2		
10	5.1		
25	4.6		
50	4.5		
100	2.4		

channel 2 were conditioned with the same constant voltage of 459 0.5 V. The output of each channel was sampled at a frequency 460 of 1 kHz. In Fig. 8, a period of the conditioning signals in 461 the case of a signal frequency of 1 Hz and the corresponding 462 outputs for channels 1 and 2 are shown. 463

For each frequency considered, the fast Fourier transform (FFT) of both outputs was calculated, and the ratio of the amplitudes at the frequency considered was assessed. In Fig. 9, an example of this evaluation was shown considering a signal frequency of 50 Hz. 468

The ratio between the FFT amplitudes of the two channels 469 Vout at different input signal frequencies is summarized in 470 Table III. 471

As can be seen from the results, the conditioning signal in a given channel does not influence the behavior of the circuit in another channel, as the ratios in Table III exceed two orders 474 of magnitude for each considered frequency.

B. Case of Study: Simultaneous Detection of IgG and IgA Anti-Tissue Transglutaminase Antibodies

To further demonstrate the behavior of the proposed mul-478 tichannel potentiostat, the device was tested on the semi-479 quantitative detection of the anti-tTG antibodies directed 480

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Fig. 8. Input configuration and output measurement in (a) channel 1 and (b) channel 2, for crosstalk evaluation.



Fig. 9. FFT of the $V_{out}(t)$ at channels 1 and 2 when a sinusoidal signal with a frequency of 50 Hz is applied to the channel 1.

⁴⁸¹ against the transglutaminase enzyme in its open confor-⁴⁸² mation (Open-tTG). To this end, a previously optimized ⁴⁸³ protocol [21] was used for the functionalization of gold



Fig. 10. (a) Prototype of the device designed and used for the tests performed for the semiquantitative analysis of IgG and IgA of the anti-tissue transglutaminase antibodies for celiac disease. (b) Schematic of the operations flow with images of the cloud visualizations.

nanoparticle-modified screen-printed electrodes (DropSens 484 DRP-110GNP, Metrohm). In particular, the Open-tTG¹ 485 (Zedira) enzyme was chemisorbed on gold nanoparticles, 486 thus allowing the recognition of anti-tTG antibodies by the 487 immobilized enzyme receptor. Detection was achieved using 488 secondary antibodies labeled with alkaline phosphatase, capa-489 ble of selectively binding to IgG or IgA anti-tTG antibod-490 ies (Thermo Fisher Scientific). After the addition of the 491 nonelectroactive hydroquinone diphosphate (Metrohm) sub-492 strate, the enzymatic reaction yields the electroactive hydro-493 quinone, the oxidation of which generates the signal output. 494 To test the performance of the multichannel device, positive 495 and negative controls of the ZediXclusive Open tTG¹-Ab 496 (IgA/IgG) ELISA kits were used. These standard solutions 497 have a concentration, respectively, over and below the thresh-498 old limit of 3 AU/mL for anti-tTG antibodies concentration. 499 These are recognized as specific biomarkers for celiac disease 500 [37]. Both IgA and IgG have to be monitored to avoid possible 501 false negative responses in the case of IgA deficiencies. 502 Considering that at least two replicates are required for each 503 analysis, the use of the proposed four-channel potentiostat 504 allows the entire analysis to be carried out with a single 505 parallel measurement, reducing the overall operation time. 506

Three replicates of positive and negative controls were 507 tested with the proposed device. In Fig. 10(a), the prototype of 508

¹Registered trademark.

the device designed and used for the tests is shown. It is worth
noting that, in this prototype version, commercial-off-the-shelf
connectors have been used. In a future engineered version,
these components can be packed together, reducing the overall
dimensions of the device, without affecting its functionalities,
as they are only adapters without any circuitry in them.

In Fig. 10(b), a schematic of the operation flow is shown. Once the data have been acquired by the device, they are sent to the cloud service for storage and sharing. The cloud service used is ThingSpeak [38], but other platforms can be exploited as well. Data can be accessed for standard web browsers from PC or mobile devices without the need for dedicated software or APP.

The four electrochemical cells were conditioned with the 522 same parameters: after 3 min when the screen-printed elec-523 trodes were left floating to allow for the enzymatic reaction 524 to occur and generate the electroactive hydroquinone, a V_{bias} 525 voltage ranging from -0.2 to 0.2 V was applied. To this 526 aim, V_Y was set at the constant voltage of 1 V, while a 527 variable voltage between 1.2 and 0.8 V was forced at the 528 RE pin through V_X . According to the DPV theory [30], the 529 resulting conditioning voltage should be a staircase waveform 530 with an increasing mean value [30]. It is worth to be noted 531 that the DPV technique has been used in these experiments. 532 but other techniques like chronoamperometry (CA) or cyclic 533 voltammetry (CV) are supported as well. Indeed, CA is based 534 on the application of fixed voltage and measurement of current 535 versus time, and the amplitude of the generated currents is 536 similar to the DPV case [29]. Regarding the CV, the triangular 537 voltage waveform required to bias the cell can be generated by 538 the 16-bit DAC independently for each channel. Furthermore, 539 the reading channel based on the combination of the TIA 540 and the proposed BLS, drives the ADC with a signal always 541 centered at the midpoint of the ADC range. Thus, both positive 542 and negative currents from the WE pin can be properly 543 converted and amplified. 544

Thanks to the 16-bit DAC, it was possible to set the 545 parameter of the conditioning voltage as those are normally 546 used on benchtop instruments (e.g., AUTOLAB PGSTAT 204 547 [13]), obtaining 319 measurement points. In particular, the 548 pulse amplitude was set to 50 mV, the step of the pulse low 549 level to 5 mV, the pulse duration to 10 ms, and the time 550 between pulses to 200 ms [30]. A preconditioning time of 30 s, 551 when the cell was kept at -0.2 V, was introduced to precon-552 centrate the reduced form of hydroquinone, thus increasing the 553 sensitivity of the analysis. To control the time intervals, the μ C 554 internal timers were exploited [31]. This ensures a total sample 555 acquisition time, for both IgG and IgA of 4'17" with two 556 replicates against the 17'10" that would occur using only one 557 channel and performing the measurements in sequence. The 558 last case was computed without considering the time needed 559 to change the electrodes at the input of the device. 560

Differential current waveforms are obtained by subtracting from each other the measured currents at the beginning and at the end of each pulse of the conditioning voltage, as required by the DPV technique. The signals recorded for the three-replicate for both IgG and IgA antibodies are reported in Fig. 11(a) and (b), respectively. Since the peak of



Fig. 11. Positive and negative data acquired for (a) IgG and (b) IgA. Positive samples are drawn with a blue line, and negatives with a red one.

the differential current is related to the concentration of the analyte, it is possible to label a sample as positive or negative through comparison with a predefined threshold. 569

From the recorded signals, the well-known baseline wan-570 dering phenomenon is evident. To overcome this and correctly 571 estimate the current peak, a baseline correction algorithm was 572 performed on the μ C platform after the signal acquisition 573 (Fig. 12). The actual baseline was estimated by computing 574 a linear interpolation of the ten first points of the differential 575 current (i.e., blue line in Fig. 12) plus ten relative minimum 576 points. The red line in Fig. 12 represents the baseline, and 577 the blue dots are the point exploited for computing it. The 578 current peak was then computed as the maximum value of the 579 distance between the differential current and the baseline. 580

As can be observed in Fig. 11, for both IgA and IgG the output signals acquired for the negative controls (i.e., red lines) are negligible, corresponding to current peaks within hundreds of nanoamperes, while intense peaks are recorded for the positive controls (i.e., blue lines).

The boxplot diagrams of the computed current peaks for negatives and positives for IgG and IgA antibodies are reported in Fig. 13(a) and (b), respectively. Again, the clear distinction

Device	Channels	Parallel channels	Max current range (resolution)	Min current range (resolution)	Voltage range (resolution)	Power Consumption per channel	Interface
[23]	32	4	±1.5µA	±1.650nA	±4V	-	Wi-Fi
		(2 electrodes		(125pA)	(2mV)		Ethernet
		cell)					
[24], [25]	128	Time	±3.3µA	-	$\pm 10V$	26.7 mW	UART
		multiplexed, 8	(100pA)		(305µV)		
		independent REs					
[27]	6	Shared CE,	-	±180nA	±5V	-	Bluetooth
		RE		(5.5pA)	(153µV)		
[12]	2	Shared CE,	$\pm 5 \text{mA}$	100nA	-2V÷2.3V	-	Bluetooth
		RE	(300nA)	(6pA)	(537µV)		
[13]	8	Shared CE,	$\pm 80 \text{mA}$	±1nA	±4V	-	Bluetooth
		RE	(40µA)	(1pA)	(960µV)		
[28]	3	1 (sequential	±500µA		±1.5V	-	USB
		sampling)			(700µV)		Bluetooth
Proposed	4	4	±62µA	620nA	±1.65V	23.5 mW	Wi-Fi
			(7nA)	(75pA)	(50µV)		

TABLE IV



Fig. 12. Baseline correction algorithm with current peak computation for a negative sample (top) and a positive sample (bottom). The red line represents the baseline computed with the linear interpolation of 20 points (marked as blue dots). The actual peak is computed as the maximum value of the distance between the baseline and the differential current (blue line).

between positive and negative controls for both IgA and IgG anti-tTG antibodies is evidenced in the boxplot charts, where a significant difference (p < 0.001) was observed.

592 C. Comparison With Other Works

The comparison of the proposed multichannel potentiostat with other previously described devices is shown in Table IV. As can be seen, the developed device is equipped with four truly independent channels, with simultaneous acquisition and independent conditioning voltages. The Wi-Fi connection,

thanks to a longer range than the Bluetooth radio, allows 598 improving portability without the need for an external device 599 nearby, acting as a gateway to the internet. Only a standard 600 Wi-Fi router, which is usually already present in a point of care 601 (PoC) or home environment, is needed to upload data to the 602 cloud. The Wi-Fi link is also exploited in [24]. In that solution, 603 the device was, however, connected to a local PC instead of a 604 cloud service, thus severely limiting the simultaneous sharing 605 of the results with multiple users. It is also worth noting that 606 the system described in [24] is based on a two-electrode cell, 607 waiving the protection of the RE against possible changes in 608 the WE potential [7]. 609

Furthermore, as it can be seen in Table IV, the maximum 610 current range of the proposed device is higher than other 611 devices presented in the literature. The commercial Sensit-612 BT [12] from PalmSens has a higher maximum current range, 613 but it has only two channels. Also, the commercial solution 614 from Metrohm [13] has some advantages in terms of the 615 number of channels and current ranges, but it requires a PC 616 and proprietary software for data processing and visualization. 617 Finally, the work presented in [28] has a higher current 618 range, but it features a shared ADC for the three channels, 619 so the sampling is sequential and not simultaneous. Moreover, 620 it requires a USB or Bluetooth communication to operate 621 and connect to a desktop or smartphone app for processing 622 and local visualization of the results. Regarding the power 623 consumption, most of the works considered do not report this 624 data. Our solution performs better with respect to [26]. The 625 power consumption of [12] is not reported; battery life is 626 reported instead; however, the battery capacity and a detailed 627 description of the measurement conditions are unknown. They 628 reported a battery life of 12 h at maximum power consumption. 629

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Fig. 13. Boxplot of the current peak of positive samples (blue) and negative samples (red) for the detection of the (a) IgG and (b) IgA antibodies.

For the sake of comparison, we can consider the worst case 630 in which continuous measurements are performed and the 631 microcontroller is always active with no power reduction 632 techniques (e.g., sleep modes management [39]) implemented, 633 and the network processor is idle and connected. Given two 634 1.5 V, 2700 mAh standard AA batteries, a battery life of 635 95 h is reached. Moreover, also considering the contribution 636 of the transmission, in the unrealistic case of continuous data 637 transmission, the battery life is reduced to 15 h, which is, in 638 any case, better than the performance reported for the Sensit-639 BT [12]. 640

V. CONCLUSION

In this article, a multichannel potentiostat for electrochem-642 ical analysis with four truly independent channels that can 643 be individually conditioned has been presented. The device 644 is compact and portable, with limited power consumption 645 (23.5 mW), and capable of both onboard processing and 646 communication over a Wi-Fi protocol to eliminate the need 647 for an external device nearby for data processing, viewing, and 648 sharing. The maximum measurable current range is $\pm 62 \ \mu A$, 649 with a resolution of 7 nA. The sensitivity is automatically 650 tuned, defining the current range during the data acquisition 651

through a multiplexer and selecting the best gain of the 652 transimpendance amplifier. The channel-to-channel mismatch 653 has been evaluated, resulting in a maximum relative error in 654 the gain of 1% when the maximum current range is selected. 655 The channel crosstalk has been demonstrated to be negligible. 656 The device shows characteristics that make it usable for 657 different types of electrochemical analysis and then suitable 658 for a large variety of contexts. As a case study, the device 659 was applied for the parallel acquisition of two replicates of 660 IgG and IgA anti-tissue transglutaminase antibodies showing 661 analytical performance fulfilling the diagnostic purposes aimed 662 at evaluating the onset of celiac disease. In this analysis, 663 a reduction of the acquisition time of 76% with respect to 664 the same measurements performed using only a channel is 665 experienced. 666

In comparison with other commercial devices or published works, the proposed device, while maintaining compatible electrical characteristics, has good portability and low power consumption that makes it suitable for use outside laboratories in home and PoC contexts. 671

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