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A stand-alone portable potentiostat with parallel channels for smart electrochemical analyses / Boni, A.; Bianchi, V.; Fortunati, S.; Giannetto, M.; Careri, M.; De Munari, I.. - In: IEEE TRANSACTIONS ON INSTRUMENTATION AND MEASUREMENT. - ISSN 0018-9456. - 72:(2023), pp. 7500112.1-7500112.12. [10.1109/TIM.2022.3228004]

Availability: This version is available at: 11381/2936644 since: 2024-05-20T07:40:45Z

Publisher: Institute of Electrical and Electronics Engineers Inc.

Published DOI:10.1109/TIM.2022.3228004

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

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Andrea Boni⁶, *Menber, IEEE*, Valentina Bianchi⁶, *Serior Member, IEEE*, Simons Eutomail⁶, Menchi Maria Carer¹⁶, *serior Member, IEEE*, Simons Eutomail⁶, *Serior Member, IEEE*, Simons Eutomail Maria Care (*Mexic Abstract***— In recent years portable potentiostats have increased in popularity allowing to perform electrochemical analyses outside the laboratories, moving them to home or point of care (PoC) environments. In this context, the idea of performing multiple acquisitions at the same time is particularly attractive to deal with replicates or with simultaneous multiple quantitative assessments of different analytes, shortening the time required for the analyses and/or increasing data reliability. Multiple parallel channels on a compact and wireless device can maximize the overall system usability. In this article, a multichan- nel Wi-Fi-based portable potentiostat is described. The device features four independent channels, which can be conditioned with different voltages. The device was designed to minimize the component count and the power consumption, obtaining 23.5 mW per channel. The system was electrically characterized, obtaining** ¹⁶ an excellent linearity ($R^2 = 0.9999$), a maximum channel-to-
¹⁷ channel mismatch of 1% when the maximum current range is **channel mismatch of 1% when the maximum current range is selected, and a negligible crosstalk among the channels. Moreover, the multichannel potentiostat was tested in the real case scenario of a semiquantitative evaluation of the anti-tissue transglutami- nase target antibodies in celiac disease. Two replicates of IgG and IgA were simultaneously acquired, showing a good capability of separating the positive and the negative samples, with a reduction of the acquisition time of 76% with respect to a single channel solution.**

²⁶ *Index Terms***— Chemical sensors, electrochemical devices, multichannel, potentiostat, Wi-Fi sensors, wireless sensors.** ²⁷

28 I. INTRODUCTION

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

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Digital Object Identifier 10.1109/TIM.2022.3228004

With reference to this, the main challenge tackled by the 38 researchers is how to implement a compact portable device 39 with cheap components and reduced power consumption without compromising the quality of the tests. It is worth noting 41 that the overall portability of the device strongly depends ⁴² also on the architecture of the system. In particular, for the 43 development of a completely stand-alone instrument, it is 44 preferable to avoid the use of additional devices for data ⁴⁵ elaboration and storage. Recently, several portable solutions 46 have been commercialized $[12]$, $[13]$, $[14]$; however, for their $\frac{47}{47}$ operation, they rely on supplementary devices such as personal ⁴⁸ computers (PCs) or tablets/smartphones connected via USB ⁴⁹ and/or Bluetooth. Also, some interesting solutions are avail-
so able in the literature $[4]$, $[15]$, $[16]$, $[17]$ with some advanced $\overline{51}$ features like the connection to a cloud service, but all require 52 the connection to an external device, limiting the portability 53 and the overall usability.

Moreover, these solutions, as most of the available portable 55 potentiostats, are designed for single analyte detection and ⁵⁶ are equipped with a single analog front end (AFE) to read 57 and control the electrochemical sensor $[4]$, $[15]$, $[16]$, $[17]$, 58 [18]. Many of these electrochemical acquisitions are timeexpensive, with a single experiment easily lasting several ϵ_0 minutes. Moreover, replicates are needed to increase the result 61 precision to obtain statistically relevant results. Furthermore, 62 sensor redundancy allows the implementation of advanced 63 fault-tolerant algorithms and data-averaging techniques that 64 can improve system robustness, efficiency, and accuracy ⁶⁵ [19]. Bioanalytical, pharmaceutical, and clinical applications 66 require simultaneous electrochemical detection that cannot 67 be obtained using a single-channel instrument. For example, 68 parallel measurements of multiple cancer markers are use- 69 ful for increasing the efficiency of cancer diagnostics and 70 therapy monitoring [20], as well as for considering possible $\frac{71}{10}$ correlations between compounds [9]. Therefore, a potentiostat $\frac{72}{2}$ with multiple acquisition channels would significantly increase $\frac{73}{2}$ the efficiency of the experiments, implementing their parallel $_{74}$ execution. The security of the

In this article, a multi-channel portable potentiostat with $\frac{76}{6}$ three electrodes for each channel suitable for both quantitative $\frac{77}{2}$ and semiquantitative electrochemical analysis, usable in a large $\frac{78}{8}$ variety of out-of-lab applications, is presented. The device $\frac{79}{60}$ features four completely independent and parallel channels, 80 capable of simultaneously managing, with different voltages, 81 four commercial screen-printed electrodes, each of which 82

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

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

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

115 II. RELATED WORKS

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

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

e introduction and the basic of mathims and the set in the same (e.g., DC, Moreover, mated and the two characteristics are a specific of the same and the sa 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.

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. The contract of the contr

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.

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

196 III. SYSTEM DESIGN

 $\frac{1}{2\pi} \int_{0}^{\frac{\pi}{2}} \frac{\sqrt{1-\frac{1}{2}}\sqrt{1-\frac{1}{2}}\sqrt{1-\frac{1}{2}}\sqrt{1-\frac{1}{2}}\sqrt{1-\frac{1}{2}}\sqrt{1-\frac{1}{2}}\sqrt{1-\frac{1}{2}}\sqrt{1-\frac{1}{2}}\sqrt{1-\frac{1}{2}}\sqrt{1-\frac{1}{2}}\sqrt{1-\frac{1}{2}}\sqrt{1-\frac{1}{2}}\sqrt{1-\frac{1}{2}}\sqrt{1-\frac{1}{2}}\sqrt{1-\frac{1}{2}}\sqrt{1-\frac{1}{2}}\sqrt{1-\frac{1}{2}}\sqrt{1-\frac{1}{$ The design of the proposed four-channel potentiostat was optimized for power consumption and component count. To this aim, devices available in the dual or quad con- figuration were preferred to maintain the same number of components and the same board size of the single-channel implementation [29]. Dedicated ADCs were introduced to improve the quantization error with respect to the A/D con-²⁰⁴ version channel available in the μ C and to implement the simultaneous sampling over the four channels. The analog components were selected for the minimum supply current and with dynamic performance (i.e., bandwidth and conversion speed) compatible with the target configuration (i.e., battery powered) and the frequency of the voltage signal and the output current [29], [30].

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

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

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 235 a standard I2C bus. 236

The AD8608 quad opamp exhibits an input current within 237 tens of picoampere, not to perturb the chemical reaction at the ²³⁸ RE pin, and an adequate output current capability to guarantee 239 a short pre-conditioning time of the chemical cell [33]. ²⁴⁰

The TIA opamp A2 is a critical device since it sets the $_{241}$ equivalent input noise current of the readout channel. Furthermore, 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 α opamp is shown in Fig. 2. α

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

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

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

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

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 μ C 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

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.

²⁷⁶ than 45

$$
GBW_{A2} > \frac{C_{RW}R_H}{2\pi (C_T R_T)^2}.
$$
 (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.

²⁸² In the design of the readout channel of the current flowing 283 into the WE terminal (I_W) , the signal swing at the TIA output ²⁸⁴ should be carefully considered. Indeed, the reading range of ²⁸⁵ the potentiostat cannot exceed the boundaries set by the output ²⁸⁶ 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 292 of the ADC, assumed from 0 to V_{ADC}

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

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

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

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

308 where N_B is the nominal resolution of the ADC, ΔI_W is the 309 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 ³¹¹ waveform.

 312 As shown in Fig. 3(b), the mismatch between V_Y , $V_{\text{M-TIA}}$, 313 and the midpoint of the ADC range prohibits achieving the

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

Fig. 2. The second is the boxing and Content in the second of the second of R_{12} and R_{23} and R_{33} and R_{42} and R_{53} and R_{64} and R_{65} and R_{66} and R_{67} and R_{68} and R_{68} and R_{68} and 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 $\frac{322}{2}$ 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 325 voltage, corresponding to the ADC input signal, is centered at 326 the V_{MID} voltage value and amplified by G $\frac{327}{2}$

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

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 331 be either limited by the TIA or the ADC, depending on the 332 value of *G* and V_Y . If the ADC saturation occurs before the \sim 333 TIA-opamp saturation, the reading range is ADC-limited, as in 334 cases 2 and 4 in Fig. $3(b)$. Depending on the setting of V_Y with 335 respect to *V*_{M−TIA}, the condition for ADC-limited range is 336

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

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

At the measurement startup, the μ C will set the TIA gain at 339 the maximum value. During the measurement, the gain will be 340 decreased as soon as the ADC output approaches the upper or 341 the lower saturation condition, corresponding to conditions (8) 342 and (9) , respectively. 343

If neither condition (8) nor (9) is fulfilled, the reading range 344 is TIA-limited. In this case, the μ C will decrease the gain 345 during the measurement at the occurrence of the following 346 condition for the ADC output code D_{O-ADC} : 347

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

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.

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

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

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

EXAMPLE 18
 EXAMPLE 18 In the present design, we used the LTC1408 device inte- grating six A/D conversion channels. The device was selected for the simultaneous sampling capability, the low power consumption, and the 14-bit nominal resolution, which is 357 suitable for the present potenstiostat [36]. The μ C provides the sampling and the clock signals to the ADC through a three-wire serial interface. The same interface allows the acquisition of the output data stream. It is worth noting that the outputs of the four channels have been connected to the same ADC. Hence, the sampling of the outputs of all the channels happens simultaneously. The results are then transmitted to the microcontroller with a serial interface, but this does not affect the timing of the measurements since the transmission is completed between two subsequent measurements.

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

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

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

TABLE I CURRENT RANGE, RESOLUTION, AND INPUT NOISE

${\bf R}_{\rm T}$	$\pm \Delta I_{\rm W}/2$	Resolution	I IN-N PMS	
$\left(\mathbf{k}\mathbf{\Omega }\right)$	(µA)	(nA)	(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		

be considered. Hence, the total current required for both the 388 analog and digital parts is 7.1 mA .

The reading range, the resolution, and the simulated ³⁹⁰ 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 392 maximum, i.e., 1 V/mA, TIA gain are reported in Table I. 393

IV. RESULTS AND DISCUSSION 394

A. System Characterization 395

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 $\frac{400}{200}$ 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. $\frac{403}{2}$

Three different values of R_{RW} were taken into account (i.e., $\frac{404}{404}$ 10 k Ω , 100 k Ω , and 1 M Ω). This allows considering different 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 V_{out} , 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 .

This analysis was repeated for each possible value of R_{RW} , 417 evaluating the maximum error in the measurement of the ⁴¹⁸ output voltage (Table II). 419

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

$$
\varepsilon_s = \frac{m_{\text{max}} - m_{\text{min}}}{m_{\text{avg}}}
$$
 (13) 424

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_{max} is the maximum value of the fit curve slope, among the four-channel data, given a dummy cell resistance μ_{427} value, m_{min} is the minimum value, and m_{avg} is the averaged value of the four-channel slopes.

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

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

 where *q*max is the maximum value of the fit curve intercept, among the four-channel data, given a dummy cell resistance value, *q*min is the minimum value, and *q*avg is the averaged value of the four-channel intercepts. The overall results are reported in Table II.

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

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

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

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

TABLE III CHANNEL CROSSTALK RESULTS

Input signal frequency (Hz)	FFT amplitude ratio $(x1000)$		
	2.6		
	4.2		
10	5.1		
25	4.6		
50	4.5		
	24		

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.

For each frequency considered, the fast Fourier transform 464 (FFT) of both outputs was calculated, and the ratio of the ⁴⁶⁵ amplitudes at the frequency considered was assessed. In Fig. 9, ⁴⁶⁶ an example of this evaluation was shown considering a signal 467 frequency of 50 Hz. 468

The ratio between the FFT amplitudes of the two channels 469 *V*out at different input signal frequencies is summarized in ⁴⁷⁰ Table III. 47

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 of magnitude for each considered frequency. ⁴⁷⁵

B. Case of Study: Simultaneous Detection of IgG and IgA ⁴⁷⁶ *Anti-Tissue Transglutaminase Antibodies* ⁴⁷⁷

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

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-
 ble of selectively binding to IgG or IgA anti-tTG antibodies (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 tTG^1 -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. $\frac{506}{200}$

Three replicates of positive and negative controls were 507 tested with the proposed device. In Fig. $10(a)$, the prototype of \sim 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 same parameters: after 3 min when the screen-printed elec- trodes were left floating to allow for the enzymatic reaction to occur and generate the electroactive hydroquinone, a V_{bias} voltage ranging from -0.2 to 0.2 V was applied. To this aim, V_Y was set at the constant voltage of 1 V, while a variable voltage between 1.2 and 0.8 V was forced at the RE pin through V_X . According to the DPV theory [30], the resulting conditioning voltage should be a staircase waveform with an increasing mean value [30]. It is worth to be noted that the DPV technique has been used in these experiments, but other techniques like chronoamperometry (CA) or cyclic voltammetry (CV) are supported as well. Indeed, CA is based on the application of fixed voltage and measurement of current versus time, and the amplitude of the generated currents is 537 similar to the DPV case [29]. Regarding the CV, the triangular voltage waveform required to bias the cell can be generated by the 16-bit DAC independently for each channel. Furthermore, the reading channel based on the combination of the TIA and the proposed BLS, drives the ADC with a signal always centered at the midpoint of the ADC range. Thus, both positive and negative currents from the WE pin can be properly converted and amplified.

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

 Differential current waveforms are obtained by subtract- ing 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 567 analyte, it is possible to label a sample as positive or negative $\frac{1}{568}$ through comparison with a predefined threshold.

From the recorded signals, the well-known baseline wandering phenomenon is evident. To overcome this and correctly $\frac{571}{200}$ 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 points. The red line in Fig. 12 represents the baseline, and the blue dots are the point exploited for computing it. The 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 581 output signals acquired for the negative controls (i.e., red ⁵⁸² lines) are negligible, corresponding to current peaks within 583 hundreds of nanoamperes, while intense peaks are recorded ₅₈₄ for the positive controls (i.e., blue lines).

The boxplot diagrams of the computed current peaks for 586 negatives and positives for IgG and IgA antibodies are reported $\frac{1}{587}$ in Fig. $13(a)$ and (b), respectively. Again, the clear distinction $\frac{1}{588}$

$[23]$ [24], [25] $[27]$	32 128	4 (2 electrodes cell) Time	(resolution) $\pm 1.5 \mu A$	$\pm 1.650nA$ (125pA)	±4V		Wi-Fi
					(2mV)		Ethernet
					$\pm 10V$	26.7 mW	UART
		multiplexed, 8	$\pm 3.3 \mu A$ (100pA)		$(305\mu V)$		
		independent					
	6	REs Shared CE,		$\pm 180nA$	$\pm 5V$		Bluetooth
		RE		(5.5pA)	$(153\mu V)$		
$[12]$	$\overline{2}$	Shared CE,	$\pm 5 \text{mA}$	100nA	$-2V+2.3V$		Bluetooth
		RE	(300nA)	(6pA)	$(537\mu V)$		
$[13]$	$\,$ 8 $\,$	Shared CE,	± 80 mA	$\pm 1nA$	±4V		Bluetooth
		RE	$(40\mu A)$	(1pA)	$(960\mu V)$		
$[28]$	3	1 (sequential	$\pm 500 \mu A$		$\pm 1.5V$		USB
Proposed	$\overline{4}$	sampling) 4	$\pm 62\mu A$	620nA	$(700\mu V)$ $\pm 1.65V$	23.5 mW	Bluetooth Wi-Fi
			(7nA)	(75pA)	$(50\mu V)$		
\times 10 ⁻⁶ Current (A) $\overline{2}$		1.4417e-07				thanks to a longer range than the Bluetooth radio, allow improving portability without the need for an external devic nearby, acting as a gateway to the internet. Only a standar Wi-Fi router, which is usually already present in a point of car	
$\mathbf 0$						(PoC) or home environment, is needed to upload data to th cloud. The Wi-Fi link is also exploited in [24]. In that solution	
-0.2	-0.1	0 0.1 Voltage (V)	0.2			the device was, however, connected to a local PC instead of	
\times 10 ⁻⁶						cloud service, thus severely limiting the simultaneous sharin	
		3.5296e-06				of the results with multiple users. It is also worth noting that	
5						the system described in [24] is based on a two-electrode cel	
Current (A)				the WE potential [7].		waiving the protection of the RE against possible changes i	
$\mathbf{0}$						Furthermore, as it can be seen in Table IV, the maximur	
-0.2	-0.1	$\mathbf{0}$ 0.1	0.2			current range of the proposed device is higher than other	
		Voltage (V)				devices presented in the literature. The commercial Sensit	
		Fig. 12. Baseline correction algorithm with current peak computation for a				BT [12] from PalmSens has a higher maximum current range	
		negative sample (top) and a positive sample (bottom). The red line represents the baseline computed with the linear interpolation of 20 points (marked as				but it has only two channels. Also, the commercial solutio	
		blue dots). The actual peak is computed as the maximum value of the distance				from Metrohm [13] has some advantages in terms of th	
		between the baseline and the differential current (blue line).				number of channels and current ranges, but it requires a P	
				Finally the work presented in [28] has a higher currer		and proprietary software for data processing and visualization	

TABLE IV PERFORMANCE COMPARISON

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).

⁵⁸⁹ between positive and negative controls for both IgA and IgG ⁵⁹⁰ anti-tTG antibodies is evidenced in the boxplot charts, where 591 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, $\frac{603}{2}$ the device was, however, connected to a local PC instead of a \sim 604 cloud service, thus severely limiting the simultaneous sharing $\epsilon_{0.05}$ of the results with multiple users. It is also worth noting that \cos 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 $[7]$.

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, $\epsilon_{0.00}$ 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 $\epsilon_{0.85}$ 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. ϵ_{29} $\times 10^{-6}$

 $\overline{7}$

6

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

⁶⁴¹ V. CONCLUSION

 In this article, a multichannel potentiostat for electrochem- ical analysis with four truly independent channels that can be individually conditioned has been presented. The device is compact and portable, with limited power consumption (23.5 mW), and capable of both onboard processing and communication over a Wi-Fi protocol to eliminate the need for an external device nearby for data processing, viewing, and 649 sharing. The maximum measurable current range is $\pm 62 \mu$ A, with a resolution of 7 nA. The sensitivity is automatically tuned, defining the current range during the data acquisition 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 ϵ_{600} 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 667 works, the proposed device, while maintaining compatible 668 electrical characteristics, has good portability and low power 669 consumption that makes it suitable for use outside laboratories 670 in home and PoC contexts.

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