A Resonance-Induced Sensitivity Enhancement Method for Conductivity Sensors

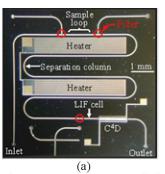
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Abstract— We demonstrated in this paper a novel technique named Resonance-Induced Sensitivity Enhancement (RISE) method to be used for significantly improving the sensitivity of a variety of conductivity sensing devices. The technique works by introducing a parallel inductor to the conductivity sensor cell and operating the sensor system at the resonant frequency of its equivalent circuit model. At the resonant frequency, parasitic capacitances that are either in series or in parallel with the target conductance can be virtually removed from the equivalent circuit and therefore a much higher sensitivity to the target conductance is obtained. In a specific example, the sensitivity of a microchip capacitively-coupled contactless conductivity detector (C⁴D) can be improved by more than 10,000 times.

I. Introduction

Conductivity sensing is a technique widely used in fields such as liquid chromatography, capillary electrophoresis, cytometry, and cell impedance analysis to analyze or detect the concentration or presence of the interested analytes [1-3]. It is often desirable to improve conductivity sensor sensitivity especially for the cases where the analytes concentrations are extremely low or the intrinsic sensor sensitivities are low due to design limitations. For example, the sensitivity of the C⁴D (capacitively-coupled contactless conductivity detector) is inferior to the conventional conductivity detector due to the fact that the sensing electrodes for C⁴D are covered by an protection layer and are not in direct contact with the electrolyte solution which means an extra serial impedance is introduced to the target impedance between the sensing electrodes [3, 4]. While the C⁴D provides great advantages such as electrode robustness, the lower sensitivity certainly limits its application and therefore improvement on this aspect is necessary.

The sensitivity degradation of C⁴D exacerbates when the sensor is built in the micro scale. For example, Fig.1 shows a homemade temperature-controlled microchip HPLC (high performance liquid chromatography) system containing a particle-packed LC (liquid chromatography) column, a 5 nL sample loop, a resistive heater, a LIF (laser-induced fluorescence) analyte detection port, and a C⁴D cell for ionic analytes detection [5]. The fabricated C⁴D cell is composed of interdigitated electrodes that are on top of the silicon dioxide layer and is further encapsulated by the parylene



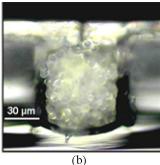


Fig. 1. Temperature-controlled microchip HPLC system. (a) Microchip top view, and (b) the cross-section of a particle-packed HPLC column.

coating as shown in Fig. 2. Silicon underneath the C⁴D electrodes was etched away by XeF_2 to create microfluidic channel cross-section [6]. Electrical routing and contact pad area outside the sensing zone was minimized to reduce the parasitic capacitance. Fig. 2(c) shows the equivalent circuit model of the C⁴D cell [3]. C_W is the capacitance between the interdigitated electrodes and the solution where the oxide/parylene layer is the capacitor dielectric. C_W was calculated to be 63 fF. R_S is the solvent resistance between the interdigitated electrodes. In general, R_S has a resistance ranging between 1 k Ω to 1 M Ω when the sensor is filled with the electrolyte solution [7]. C_P is the parasitic capacitance between the electrodes including fingers, routings, and contact pads. The capacitance of C_P was measured using the HP4192A impedance analyzer to be

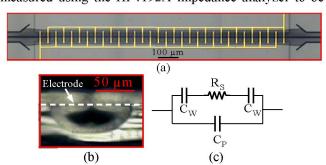
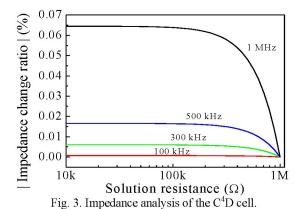


Fig. 2. C⁴D cell for analyte detection in the microchip HPLC system. (a) C⁴D cell top view, (b) C⁴D microfluidic channel cross-section, and (c) equivalent circuit model for the C⁴D cell.



1.92 pF. For high C^4D sensitivity, R_S needs to dominate the overall cell impedance. However, for the microchip C^4D system, things go the opposite way when compared with macro scale capillary C^4D [3]. In other words, R_S is small due to the short distance between electrodes; C_W is small and therefore large impedance due to the small interdigitated electrode area; C_P is large and therefore small impedance due to the large size electrical contact pads and the semi-conducting silicon substrate underneath the 1µm-thick oxide layer. Therefore, it can be expected that the microchip C^4D sensitivity to be much lower than that of the capillary C^4D or microchip conductivity sensor where electrodes are in contact with the analyte solution.

Fig. 3 shows the simulation analysis of the microchip C^4D impedance based on its equivalent circuit model and component values mentioned earlier. Results indicate that even at 1 MHz sensing frequency, the cell impedance magnitude changes by less than 0.1% when the solution resistance changes from 1 M Ω to 10 k Ω .

II. CONCEPT OF RISE METHOD

The concept of RISE (resonance-induced sensitivity enhancement) method is illustrated in Fig. 4. First, an inductor L_S with an internal serial resistance of R_{LS} is put in parallel with the C⁴D cell. In our study, a discrete inductor

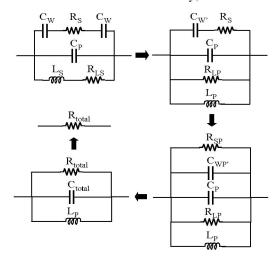


Fig. 4. Concept of the RISE method.

component was used but the inductor can as well be an integrated component fabricated on the microchip [8]. The serial circuit branch containing L_S and R_{LS} is transformed into an equivalent parallel circuit composing an inductor L_P and a resistor R_{LP} according to the following equations

$$\begin{split} Q_{LR} &= \frac{\omega_0 L_S}{R_{LS}} \\ L_P &= L_S (\frac{{Q_{LR}}^2 + 1}{{Q_{LR}}^2}) \\ R_{LP} &= R_{LS} ({Q_{LR}}^2 + 1) \end{split}$$

, where the operation frequency ω_{θ} will be defined latter. The two parylene wall capacitors C_{W} are combined into C_{W} , i.e.,

$$C_{\mathbf{W}'} = \frac{C_{\mathbf{W}}}{2} .$$

Then, the serial circuit of $C_{W'}$ and R_S is transformed into an equivalent parallel circuit composing a capacitor $C_{WP'}$ and a resistor R_{SP} according to the following equations

$$Q_{CR} = \frac{1}{R_S \omega_0 C_W},$$

$$C_{WP'} = C_W, (\frac{Q_{CR}^2}{Q_{CR}^2 + 1}).$$

$$R_{SP} = R_S (Q_{CR}^2 + 1)$$

The resistance R_{SP} and R_{LP} are combined into R_{total} and the capacitance $C_{WP'}$ and C_P are combined into C_{total} , i.e.,

$$\begin{split} R_{total} &= R_{\rm SP} \, / / \, R_{\rm LP} \\ C_{total} &= C_{\rm P} + C_{\rm WP}, \end{split} \label{eq:reconstruction}$$

Now, the operation frequency ω_{θ} is chosen so that C_{total} and L_P reaches resonance and the cell impedance becomes a pure resistance R_{total} , where

$$\begin{split} & \omega_{0} = \frac{1}{\sqrt{2}} (\sqrt{\frac{1}{C_{p}C_{W}^{2}L_{z}^{2}R_{z}^{2}} (-C_{p}L_{z}^{2} - C_{W}L_{z}^{2} + C_{W}^{2}R_{z}^{2}(L_{z} - C_{p}R_{Lx}^{2}) + }{\sqrt{-4C_{p}C_{W}^{2}L_{z}^{2}R_{z}^{2}(-L_{z} + (C_{p} + C_{W})R_{Lx}^{2}) + (C_{w}L_{z}(L_{z} - C_{W}R_{z}^{2}) + C_{p}(L_{z}^{2} + C_{w}^{2}R_{z}^{2}R_{Lx}^{2}))^{2}}}) \\ & R_{boul} = \frac{(R_{Lx}^{2} + L_{z}^{2}\omega_{0}^{2})(1 + C_{w}^{2}R_{z}^{2}\omega_{0}^{2})}{R_{Lx} + C_{w}^{2}R_{x}R_{x}^{2}R_{x}^{2}(R_{y}^{2} + C_{w}^{2}R_{x}^{2}R_{x}\omega_{0}^{2}) + C_{y}^{2}L_{x}^{2}R_{x}^{2}\omega_{0}^{2})} \end{split}$$

In order to have R_{total} strongly dependent on the solution resistance R_S , discrete component values, L_S and R_{LS} , should be chosen in a way that R_{LP} is much larger than R_{SP} so that R_{total} is dominated by R_{SP} which in turn has a strong dependence on R_S .

III. EXPERIMENTAL RESULTS

As a first step to examine the performance of the RISE technique, a set of inductor component values were chosen where L_S is 15 mH and R_{LS} is 30 Ω . These component values were extracted from a discrete coil inductor using the impedance analyzer. The microchip C^4D component values are the same as mentioned earlier, i.e., $C_W = 63$ fF and $C_P = 1.92$ pF. Using those component values, HSPICE analysis of the circuit was carried out. Fig. 5 then shows the analysis results which are frequency scans of the cell impedance magnitude. Different curves were plotted for different solution resistance R_S which is $1 \text{ k}\Omega$, $10 \text{ k}\Omega$, $100 \text{ k}\Omega$ and $1 \text{ M}\Omega$, respectively.

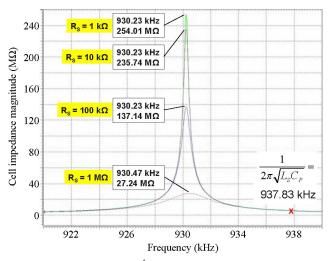


Fig. 5. HSPICE analysis of C⁴D cell impedance magnitude versus frequency.

The resonant frequencies extracted from the HSPICE results where the impedance magnitude curves reach the maxima match exactly with the calculated frequencies from the ω_0 equation. It is also clear from Fig. 5 that at the resonant frequency (930.23 kHz), cell impedance magnitude change ratio due to solution resistance change (from 1 M Ω to 1 k Ω) reaches its maximum while if operating at the resonant frequency of L_S and C_P (937.83 kHz) there is virtually no impedance magnitude change.

The operation resonant frequency remains at 930.23 kHz for a solution resistance ranging from 1 k Ω to 100 k Ω and slightly increases to 930.47 kHz where the solution resistance is 1 M Ω . Since the resonant frequency is not sensitive to solution resistance in the designated solution resistance range, we chose the operation frequency f_0 to be 930.23 kHz or ω_0 to be 5844.81 krad-Hz in our following simulation. As shown in Fig. 6, R_{total} (total cell impedance at the resonant frequency) versus solution resistance R_S curve is plotted and impressively R_{total} changes by 765% when the solution resistance changes from 1 M Ω to 10 k Ω . Compared with the native C⁴D performance demonstrated in Fig. 3, the sensitivity enhancement by RISE method is more than 10.000 times.

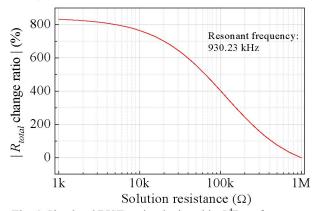


Fig. 6. Simulated RISE-assisted microchip C⁴D performance.

TABLE I. Experimental verification of RISE method using a model circuit built with discrete components.

$C_{\rm W}$ =18.2 nF $C_{\rm p}$ = 0.1 μ F $L_{\rm S}$ = 8.64 mH $R_{\rm LS}$ = 78.8 Ω	\mathbf{f}_0 $(\boldsymbol{\omega}_0/2\pi)$	R_{total} (with RISE) $(R_{\rm S} = 1000\Omega)$		$\begin{aligned} & Z_{total} \\ &(w/o~RISE)\\ &(R_{_S} = 1000~\Omega) \end{aligned}$		Sensitivity Enhancement ratio
Theoretical values	4.98 kHz	939.7 Ω	1004.9 Ω	294.7 Ω	292.9 Ω	11.36
Experimental results	5.16 kHz	940.4 Ω	1004.7 Ω	281.8 Ω	280.0 Ω	10.70
Error	3.61%	0.07%	-0.02%	-4.38%	-4.40%	-5.81%

As a first step to experimentally verify the RISE technique, a model RISE-assisted C⁴D circuit built with discrete resistors, inductors, and capacitors was created. Component values are: $C_W = 18.2$ nF; $C_P = 0.1$ µF; $L_S = 8.64$ mH; $R_{LS} = 78.8$ Ω ; $R_S = 1$ or 1,000 Ω . R_{total} was measured for different R_S values under the resonant frequency ω_0 where the circuit impedance magnitude maximized. $|Z_{total}|$ is the measured impedance magnitude of the native C⁴D circuit (no L_S and R_{LS}) at the frequency ω_0 . The experimental results in TABLE I show great matching between theoretical and experimental values.

Furthermore, RISE technique was formally applied to our microchip C^4D device to verify the sensitivity enhancement performance. In this experiment, L_S is 32 mH and R_{LS} is 16 k Ω . As shown in TABLE II, media of different electrical conductivities (air, DI water, and 1M NaCl water solution) were sent into the C^4D microfluidic channel and the cell impedance magnitude with/without RISE assistance was recorded. The resonant frequency was experimentally measured using the HP4192A impedance analyzer to be 633 kHz. Results showed that the RISE method significantly enhanced the microchip C^4D sensitivity. The exact sensitivity enhancement ratio was not available here due to the limited resolution of the impedance analyzer.

TABLE II. Experimental verification of RISE method using the microchin C⁴D device

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$L_{\rm S} = 32 \text{ mH}$ $R_{\rm LS} = 16 \text{ k}\Omega$ $f_0 = 633 \text{ kHz}$	Medium = air	Medium = DI water	Medium = 1M NaCl water	Impedance change ratio				
Impedance magnitude (w/o RISE)	130 kΩ	129 k Ω	130 kΩ	< 1%				
Impedance magnitude (with RISE)	803 kΩ	895 kΩ	1127 kΩ	40.3%				

IV. RISE METHOD OPTIMIZATION

While experimental results from section III illustrated that RISE method is capable of providing significant sensitivity enhancement for microchip C⁴D, the RISE method can be further optimized with respect to specific sensing parameters and will be discussed as follows.

First, for conventional coil inductors, the inductance is proportional to the square of the number of coil turns N, while the internal serial resistance is proportional to the number of coil turns, i.e.,

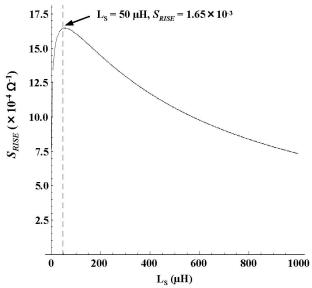


Fig. 7. RISE method optimized to S_{RISE} for R_S equal to 1 M Ω .

$$\begin{split} R_{\scriptscriptstyle LS} &\propto N \propto \sqrt{L_{\scriptscriptstyle S}} \\ or, \ R_{\scriptscriptstyle LS} &= A * \sqrt{L_{\scriptscriptstyle S}} \end{split}$$

Using one set of the measured component values ($L_S = 15$ mH and $R_{LS} = 30 \Omega$) we then get

$$A = \frac{R_{LS}}{\sqrt{L_S}} = \frac{30}{\sqrt{15E - 3}}$$

$$R_{LS} = 30\sqrt{L_{LS}/15E - 3}$$

The RISE-assisted C^4D sensitivity S_{RISE} is defined here as

$$S_{RISE} = \left| \frac{\partial R_{total}}{R_{total} \partial R_{S}} \right|.$$

Now, say if we need to optimize conductivity sensitivity for R_S around 1 M Ω , simply plot S_{RISE} versus inductance L_S with R_S equal to 1 M Ω and then locate the L_S value where S_{RISE} is maximized as shown in Fig. 7.

Another way to optimize RISE method is to maximize the C⁴D impedance change ratio γ_{RISE} when R_S changes from 1 M Ω to 1 k Ω , i.e.,

$$\gamma_{\rm RISE} = \frac{R_{total(R_{\rm g}=1M\Omega)} - R_{total(R_{\rm g}=1k\Omega)}}{R_{total(R_{\rm g}=1M\Omega)}} \times 100\% \ . \label{eq:gamma_risk}$$

By plotting γ_{RISE} versus L_{S} , the maximum ratio is found to be 2249% where L_{S} is 600 μ H. TABLE III then shows the summary of the RISE performances with and without optimization.

TABLE III. Summary of RISE method optimization.

	Without optimization	Optimized for impedance change ratio	Optimized for sensitivity	Enhancement ratio
Component values	$L_{\rm S} = 15 \text{ mH}$ $R_{\rm LS} = 30 \Omega$ $f_0 = 930.23 \text{ kHz}$	$L_s = 600 \mu H$ $R_{LS} = 6 \Omega$ $f_0 = 4.65 \text{ MHz}$	$L_{s} = 50 \mu H$ $R_{LS} = 1.73 \Omega$ $f_{0} = 16.11 \text{ MHz}$	N/A
Y _{RISE}	832%	2249%	N/A	270.3%
S_{RISE}	8.02E-5	N/A	1.65E-3	2057%

V. CONCLUSION

In this paper, we demonstrated the concept and performances of the RISE technique to significantly enhance the sensitivity of conductivity sensors. The proposed technology is efficient, low-cost and easy to implement. It is important to understand that RISE can be applied to versatile conductivity sensing applications and not just to microchip C⁴D for HPLC analyte detection. For example, RISE can be used in the conventional capillary HPLC or capillary electrophoresis (CE) systems to improve the macro-sized C⁴D sensitivity. It can also be used to enhance the sensitivity of conventional conductivity sensors where sensing electrodes are in direct contact with the electrolyte solution. In this case, it is the double-layer capacitance on top of the electrodes [7] as well as the parasitic capacitance from electrode routing that will be virtually removed from the circuit using RISE.

REFERENCES

- [1] Q. He, C. Pang, Y. C. Tai, and T. D. Lee, "Ion liquid chromatography on-a-chip with beads-packed parylene column," presented in the 17th IEEE International Conference on Micro Electro Mechanical Systems (MEMS 2004), Maastricht, The Netherlands, 2004.
- [2] P. Kuban, and P. K. Dasgupta, "Capillary ion chromatography," J. Sep. Sci., vol. 27, pp. 1441-1457, 2004.
- [3] A. J. Zemann, E. Schnell, D. Volgger, and G. K. Bonn, "Contactless conductivity detection for capillary electrophoresis," Anal. Chem., vol. 70, pp. 563-567, 1998.
- [4] J. Tanyanyiwa, and P. C. Hauser, "High-voltage capacitively coupled contactless conductivity detection for microchip capillary electrophoresis," Anal. Chem., vol. 74, pp. 6378-6382, 2002.
- [5] C. Y. Shih, W. Li, and Y. C. Tai, "A novel embedded liquid chromatography system with submicron-sized filter structure and capacitively-coupled contactless conductivity detector," presented in the 20th International Symposium on Microscale Bioseparations, Amsterdam, the Netherlands, 2006.
- [6] C. Y. Shih, and Y. C. Tai, "Single-mask technology for on-chip high-pressure HPLC system," presented in the 9th International Conference on Miniaturized Systems for Chemistry and Life Sciences, Boston, United States, 2005.
- [7] S. Zheng, M. Liu, H. L. Kasdan, and Y. C. Tai, "Platinum black electroplated impedance particle sensor," presented in the 1st Annual IEEE International Conference on Nano/Micro Engineered and Molecular Systems, Zhuhai, China, 2006.
- [8] W. Li, D. C. Rodger, E. Meng, J. D. Weiland, M. S. Humayun, and Y. C. Tai, "Flexible parylene packaged intraocular coil for retinal prostheses," presented in the 4th International IEEE-EMBS Special Topic Conference on Microtechnologies in Medicine and Biology, Okinawa, Japan, 2006.