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# CMOS Direct Time Interval Measurement of Long-Lived Luminescence Lifetimes

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*Abstract*— **We describe a Complementary Metal-Oxide Semiconductor (CMOS) Direct Time Interval Measurement (DTIM) Integrated Circuit (IC) to detect the decay (fall) time of the luminescence emission when analyte-sensitive luminophores are excited with an optical pulse. The CMOS DTIM IC includes 14x14 phototransistor array, transimpedance amplifier, regulated gain amplifier, fall time detector, and time-to-digital convertor. We examined the DTIM system to measure the emission lifetime of oxygen-sensitive luminophores tris(4,7-diphenyl-1,10- phenanthroline) ruthenium(II) ($[Ru(dpp)_3]^{2+}$) encapsulated in sol-gel derived xerogel thin-films. The DTIM system fabricated using TSMC 0.35µm process functions to detect lifetimes from 4µs to 14.4µs but can be tuned to detect longer lifetimes. The system provides 8-bit digital output proportional to lifetimes and consumes 4.5mW of power with 3.3V DC supply. The CMOS system provides a useful platform for the development of reliable, robust, and miniaturized optical chemical sensors.**

## I. Introduction

Luminescence lifetime based sensing is a good platform for the development of robust, field-usable and reliable sensors for biological, medical, and chemical sensing applications [1, 2]. Luminescence lifetime based sensors are insensitive to (a) light source (excitation) and detector drift, (b) changes in optical path, and (c) drift due to luminophore degradation and/or leaching [1, 3]. We employ a CMOS optoelectronic IC with monolithically integrated photodetection elements and signal processing circuitry to work towards low cost miniaturized sensors [4]. In a microsystem configuration, direct detection of luminescence lifetimes is challenging and less commonly pursued as it can necessitate elaborate signal detection and processing instrumentation when performed in the time-domain, because the lifetimes are relatively short and can range from several nanoseconds to milliseconds. To directly measure the luminescence lifetimes in time-domain, the sample is excited by a light pulse signal with a short pulse width and the subsequent exponentially decaying behavior of the emission response is monitored.

In the history of luminescence lifetime based sensors, Trettnak, et al. [5] first developed a miniaturized oxygen sensor instrumentation utilizing a phase modulation technique by assembling off-the-shelf integrated circuits into a compact instrument. Then, Kieslinger, et al. [6] demonstrated a compact sensor instrumentation for direct time-domain measurement of fluorescence lifetimes again based on assembling off-the-shelf integrated circuits controlled by a microcontroller. Many luminescence lifetime-based integrated sensors that utilize the phase modulation technique have also reached commercial applications such as measurement of oxygen in waste water monitoring and modified atmosphere food packaging by McDonagh, et al. and von Bultzingslowen, et al., repectively [7, 8] . More recently, NeoFox fiber-optic sensor system for biological and medical applications was introduced by Ocean Optics Inc. [9]. We were the first research group to pioneer the development of custom-designed CMOS ICs to perform phase luminometric signal detection and processing in both linear [4] and non-linear (enhanced sensitivity) [10] operations. To date, the resolution of CMOS phase luminometry systems achieved to detect lifetimes is relatively low [4] and special non-linear signal processing techniques are required to improve the phase detection resolution [10] to obtain a large voltage/current output per unit change in analyte concentrations.

Here, we propose a mixed-signal CMOS system for direct luminescence lifetime measurements using Direct Time Interval Measurement (DTIM) method which uses the strategy of using voltage comparators that trigger when the exponentially decaying luminescence response (under excitation by a light pulse) crosses specific voltage levels. The DTIM IC then uses a Time-to-Digital Convertor (TDC) circuit to measure the time internal between the triggering of subsequent voltage comparators to provide the time domain characteristics of the luminescence response. In principle, this strategy could be used to measure luminescence lifetimes that are on the order of few hundred nanoseconds. The limiting factor to detecting even shorter lifetimes is the use of large sized photodetectors to detect the weak luminescence signals which introduces significant parasitic capacitances which in turn limits the operational bandwidth. The prototype system is suitable for use with luminophores with relatively long-lived excited-state lifetimes ranging from several microseconds or longer lifetimes [1] and provides an 8-bit digital output. The sensor system block diagram is shown in Figure 1.

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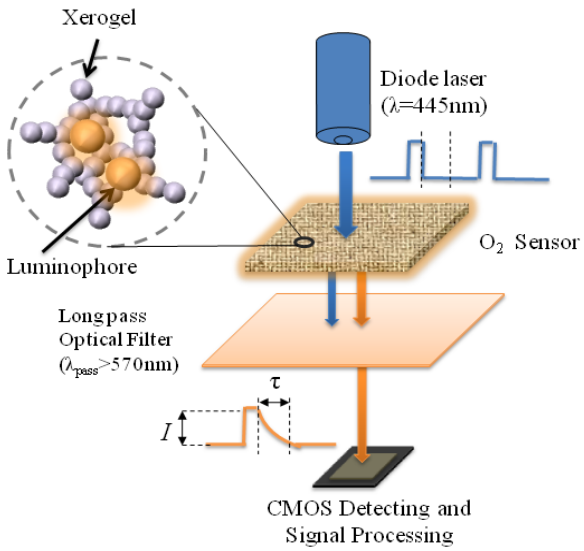


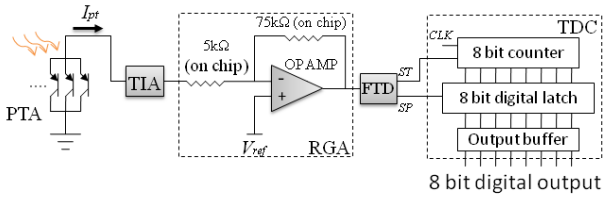

**Fig.1.** Block diagram of the CMOS Luminescence Lifetime Detection System.

We employ xerogels that are sol-gel process derived materials and that have been extensively used as immobilization media for sequestering a variety of recognition elements including fluorophores, enzymes, and modified nanoparticles [2, 11, 12]. Xerogels are nanoporous glasses and their appeal for biorecognition elements derives from their production at room temperature, thermal stability, tunable pore dimensions, biocompatibility, and a broad optical transparency window. The oxygen sensor described here is based on encapsulating the luminophore tris(4,7-diphenyl-1,10-phenanthroline) ruthenium(II) ($[Ru(dpp)_3]^{2+}$ which is well known to be responsive to gaseous oxygen, in the xerogel matrices [3]. Assuming all the luminophore molecules in the xerogel-thin-film are equally accessible to the $O_2$ molecules, Equation (1) (known as Stern-Volmer equation) describes the relationship between $O_2$ concentration and luminophore quenching by $O_2$, where $I_0$ and $\tau_0$ are the luminescence intensity or lifetime in the absence of $O_2$ respectively, I and τ are intensity or lifetime in the presence $O_2$ respectively, $Ksv$ is the Stern-Volmer constant, $k_q$ is the bimolecular quenching constant and $[O_2]$ is the gaseous $O_2$ concentration [3].

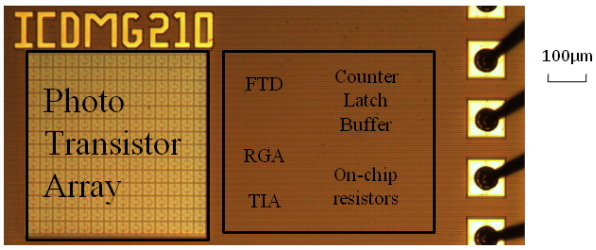
$$\frac{I_0}{I} = \frac{\tau_0}{\tau} = 1 + K_{sv}[O_2] = 1 + K_q \tau_0 [O_2] \qquad (1)$$

## II. CMOS DTIM IC

In literature, we can find several reports of CMOS optoelectronic ICs that can perform time-resolved fluorescence imaging and optical time-of-arrival measurements for many applications including DNA analysis, quantum dot imaging, pH sensing, and fluorescent dyes imaging [13-18]. These systems typically use avalanche photodiodes as detectors which require high voltage biases and are unstable with ambient temperature changes limiting their use in versatile situations. We aim to use phototransistors as detectors that provide high responsivity and can be operated with low power. The CMOS DTIM IC is designed and fabricated in TSMC 0.35µm CMOS technology and its block diagram is shown in Figure 2(a). The DTIM IC consists of five circuit components which include (i) Phototransistor Array (PTA), (ii) Transimpedance Amplifier (TIA), (iii) Regulated Gain Amplifier (RGA) (iv) Fall Time Detector (FTD), and (v) Time-to-Digital Convertor (TDC). Figure 2(b) shows the microphotograph of the CMOS IC. The circuitry (except PTA) is covered by a metal layer to prevent noise due to interference of optical signals with circuitry.




(a)




(b)

**Fig. 2.** DTIM circuit Block diagram and microphotograph of the DTIM IC. (PTA: phototransistor array, TIA: transimpedance amplifier, RGA: regulated gain amplifier, FTD: fall time detector and TDC: time-to-digital convertor).

In operation, a diode laser is used to generate short pulses repeating at a low frequency (few hundred hertz) to excite the luminophore molecules which subsequently generate exponential decaying luminescence response. The luminescence signal is made to incident on the PTA which generates a proportional photo-current signal, $I_{pt}$. The PTA consists of 196 identical phototransistors connected in parallel and arranged in 14 x 14 array to reduced the effective capacitance of the photodetector to improve the bandwidth while providing a large signal collection area. Each phototransistor is similar to a bipolar junction transistor and is formed by p-active (emitter) / n-well (base) / p-substrate (collector). We previously examined this vertical phototransistor which has high sensitivity in visible electromagnetic spectrum [4]. The photo-current, $I_{pt}$, from the PTA is converted to a voltage signal and amplified by the TIA. The output of the TIA is amplified by RGA. The gain of the RGA is set at 15 V/V using on-chip resistors. A FTD is used to detect the decay time of the signal from the RGA. The FTD consists of a series of 16 comparators that are used to generate positive edges based on the voltage level of the input signal. The output of FTD is connected to a TDC. In TDC, the positive edge of the signal ST will activate an 8-bit counter. Later, the positive edge of signal SP will latch the current output of the counter. The latched 8-bit digital output represents the time interval, τ (exponential decay of the luminescence response as shown

in Figure 1), which is read out through a buffer. The time resolution of the TDC is decided by the frequency of external clock signal, *CLK*.

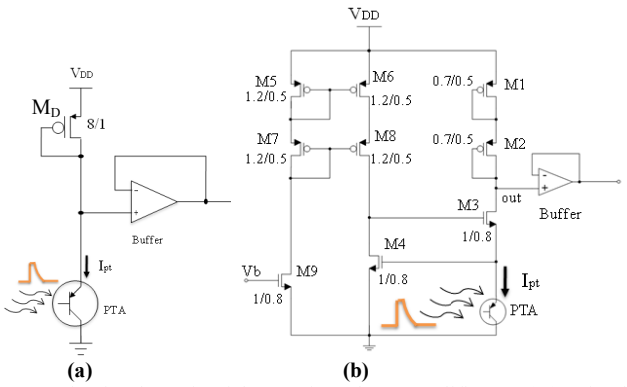



**Fig. 3.** Circuit schematic of the transimpedance amplifier (TIA) (a) Simple diode connected TIA; (b) Reduced Gate Capacitance (RGC) TIA. All mentioned W/L's of the MOSFETs are in micrometers

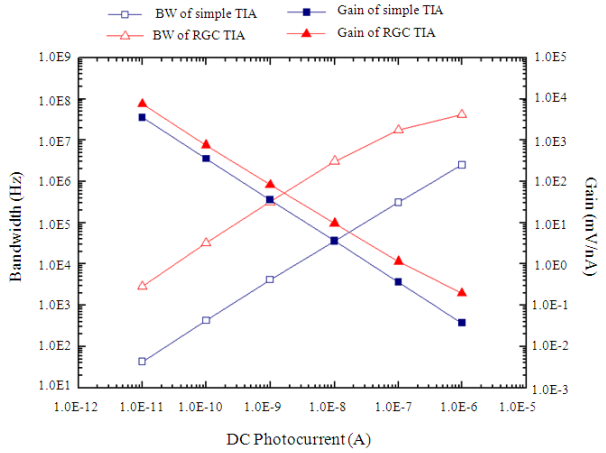



**Fig. 4.** Simulated comparison between the gain and bandwidth of the simple diode connected load TIA and RGC-TIA used in the CMOS DTIM IC by varying with DC input photocurrent level.

### *A. Transimpedance Amplifier (TIA)*

The front-end TIA is important in determining the DTIM system bandwidth. We examined the performance of two TIA circuits. The first is a simple diode connected TIA as shown in Figure 3(a). The gain and bandwidth of simple diode connected load TIA can be expressed [19, 20] as,

$$A_{TIA} = \frac{1}{g_m} = \frac{nV_T}{I_{dc}} \quad (2)$$

$$BW_{TIA} = \frac{g_m}{2\pi C_{PT}} \quad (3)$$

where, $g_m$ is the small signal transconductance of diode connected transistor, n is subthreshold slope factor of $M_D$, $V_T$ is the thermal voltage, $I_{dc}$ is the DC photocurrent and $C_{PT}$ is the parasitic capacitance of phototransistor array, PTA. To increase the bandwidth of TIA, one can either increase $g_m$ or decrease $C_{PT}$. The value of $g_m$ cannot be very large as we are detecting weak optical signals ($I_{pt}$ is very small on the order of few nano-amperes) and high gain is needed to amplify the small ac-photocurrent. Also, $C_{PT}$ is fixed as the size of the PTA is fixed (196 phototransistor pixels connected in parallel, arranged in a 14 x 14 array occupying 490µm by 490µm area). The PTA area was selected based on our previous experimental knowledge as necessary to detect the weak luminescence signals [4, 10]. Hence, we use the Reduced Gate Capacitance (RGC) TIA which effectively reduces $C_{PT}$ as seen by the TIA output node without needing to decrease the area of the PTA [19]. The circuit of the RGC-TIA as shown in Figure 3(b) is used in the DTIM IC. Specifically, the output node is buffered by M3 and M4 from the PTA. The gain and bandwidth of the RGC-TIA can be derived [19] as,

$$A_{TIA} = 2 \cdot \frac{1}{g_m} = \frac{2nV_T}{I_{dc}} \quad (4)$$

$$BW_{TIA} = \frac{g_m}{2\pi C_p}$$

$$C_p = C_{GB,M2} + C_{DB,M3} + C_{in,Buffer} \quad (5)$$

The $C_{PT}$ in Equation (3) is replaced by $C_p$ which is the joint capacitance of gate-GND capacitance of M2, drain-GND capacitance of M3 and input capacitance of the buffer. $C_p$ is designed to be on the scale of several femto-farads which is significantly smaller than $C_{PT}$, which causes a large increase in the bandwidth. Figure 4 shows the simulation comparison between the gain and bandwidth of the simple diode connected TIA and RGC-TIA used in the CMOS DTIM IC by varying with DC input photocurrent level. From Figure 4, the bandwidth of RGC-TIA is increased several times than the simple diode connected load TIA. Based on our previous knowledge, we estimate that the photocurrent generated from PTA will be on the order few nano-amperes, which will give a maximum bandwidth of 5MHz [4, 10]. Our simulations show that input photocurrent pulses with falling/rise times of 200ns can be accurately detected by the RGC-TIA.

### *B. Regulated Gain Amplifier (RGA), Fall Time Detector (FTD), Time to Digital Converter (TDC)*

A regulated gain operational amplifier is used to amplify the voltage signal from RGC-TIA. The amplifier is a standard two-stage circuit with miller compensation [21]. The voltage gain of the operational amplifier is set as 15V/V using on-chip polysilicon resistors. The bandwidth of the operational amplifier is designed as 206MHz which is larger than the bandwidth of the RGC-TIA. The output of the RGA is connected to Fall Time Detector.

The Fall Time Detector, FTD consists of a series of 16 comparators that are used to generate positive edges based on the voltage level of the input signal. There are two important issues to be considered in the design of the FTD. First, $V_{plus}$ has to automatically adjust each time as the signal changes. Second, the fall time of the comparator output should be relatively constant with respect to its input DC level. To solve the first issue, a voltage level independent FTD is designed as shown in Figure 5(a). $V_{plus}$ is fixed and

the signal voltage level varies between $V_{minus}$ and $V_{plus}$. The output of the comparator array is connected to a D Flip-Flop array and logic gates are used to track the first positive edge which indicates when the input signal starts falling. The time interval between positive edge of signal, ST and positive edge of signal, SP is the fall (decay) time of the input signal. The accuracy of this FTD is dependent on the number of comparators, and in the current prototype 16 comparators are used. Figure 5 (b) shows the typical signal waveform from the FTD. To solve the second issue discussed above, the comparator is designed using complementary input stage [21]. When the input DC level varies, the bias current of the comparator input stage remains stable because of the compensation of NMOS/PMOS input differential pair and so does the output rise/fall time. The fall/rise time of the complementary comparator stays within 10ns±6ns range through the full 0V to 3.3V input DC range.

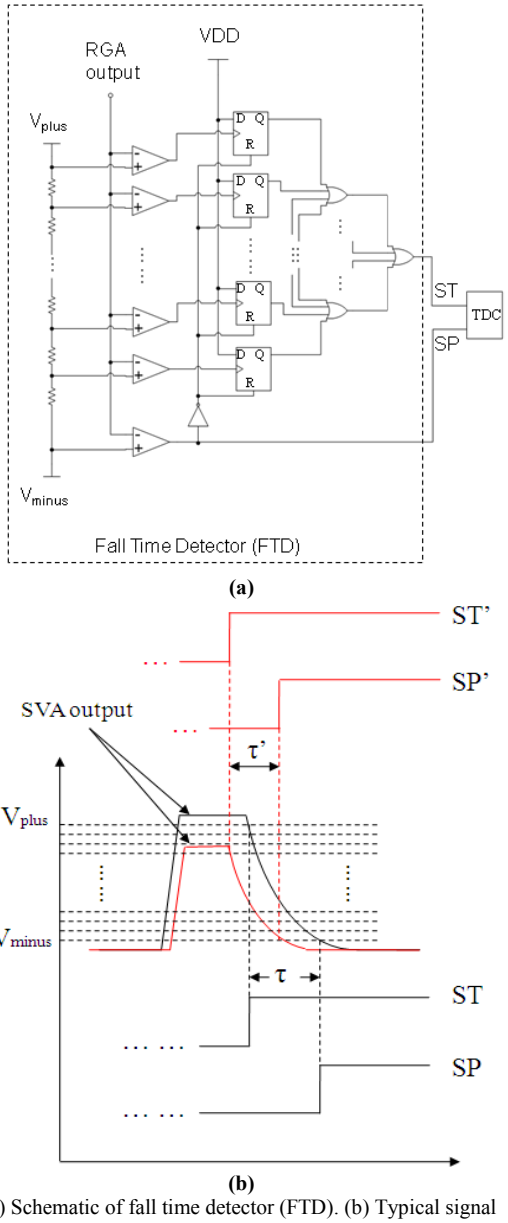



**Fig. 5.** (a) Schematic of fall time detector (FTD). (b) Typical signal waveform processing by the FTD.

The output of FTD is connected to the TDC. Figure 2(a) shows the schematic of the TDC which is built using standard digital cell library supplied by TSMC. The TDC works as follows: 1) the positive edge of signal ST will activate the counter, 2) after τ seconds the positive edge of signal SP will active the latch to fix the current output of the counter. 3) The latched 8-bit digital output representing the time interval τ is read out by other digital circuits.

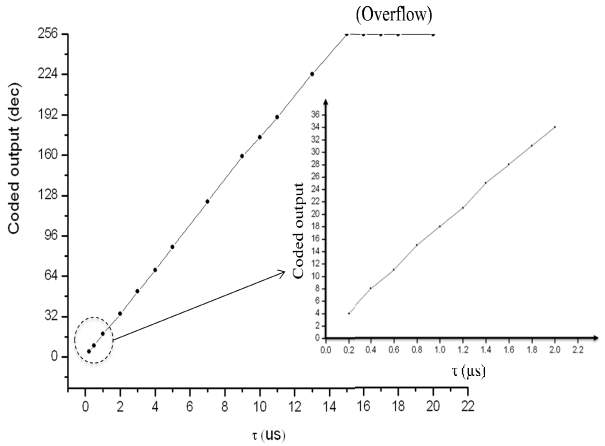



**Fig. 6.** Simulation performance of the complete DTIM IC

## III. Simulation and Experimental Characterization of the DTIM IC and Oxygen Sensor

Figure 6 shows the simulation of the complete DTIM IC. An ideal current source (pulse input with 9nA as 'low', 11nA as 'high') connected in parallel to a 1pF capacitor (estimated value of the parasitic capacitance of PTA) is given as input to RGC-TIA. The frequency of CLK in TDC is set at 20MHz for a time resolution of 50ns. From Figure 6, the relationship between the fall time of the input current pulse and the system output (8-bit digital output) is linear as the fall time is varied from 200ns to 14.4μs.

Figure 7 shows the experimental measurement result of the DTIM IC when an optical signal from a diode laser (Coherent Inc., Model: Cube λ=445nm) with 4μs fall time and optical power of 10nW is incident on the PTA. The oscilloscope view shows the output of RGA and FTD. The response time of the PTA, RGC-TIA and RGA cause the fall time to extend to 4.2μs. The fall time measured by FTD is 4μs as the triggering for ST and SP is calibrated to accommodate the response time delay of PTA, TIA and RGA, which varies little throughout the designed DTIM IC operational range. From Figure 6, the DTIM system was designed to operate with emission decay lifetimes ranging from 200ns to 14.4μs, however, our predicted value for the capacitance of the PTA, $C_{PT}$ (Equation 3 and 4) was lower than the actual value of the fabricated IC. The increased value of the $C_{PT}$ caused a reduction in the system bandwidth and in practice DTIM system was reliably able to detect lifetimes ranging from 4μs to 14.4μs. The DTIM IC consumed 4.5mW of power with 3.3V DC supply and can detect luminescence lifetimes with 50ns resolution between the operational range.

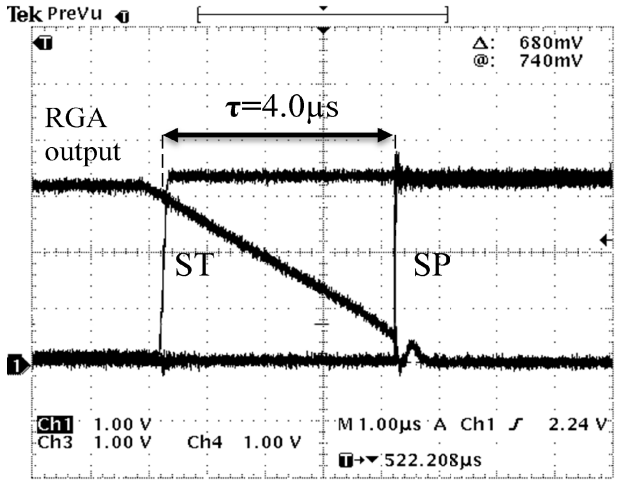



**Fig. 7.** Experimental characterization of the DTIM IC

To fabricate the xerogel based oxygen sensors, the following reagents were used: tris(4,7'-diphenyl-1,10'-phenathroline) ruthenium(II) chloride ($[Ru(dpp)_3]Cl_2$) (GFS Chemical Inc); tetraethoxysilane (TEOS) (Gelest Inc.); *n*-octyltriethoxysilane (C8-TEOS) (Gelest Inc.); hydrochloric acid (HCl) (J.T. Baker); and ethanol (EtOH) (Quantum Chemical Corp.). All reagents were used as received without further purification. A sol was prepared by mixing of TEOS (1.448 mL, 6.5 mmole), C8-TEOS (2.052mL, 6.5mmole), EtOH (2.52mL, 44mmole) and HCl (0.8mL of 0.1N HCl, 0.08mmole). This mixture was capped and magnetically stirred under ambient conditions for 1 h. 25mM $[Ru(dpp)_3]Cl_2$ (146.2mg, 0.3mmole) in ethanol (5mL, 85mmole) was prepared as a stock solution. 0.273mL of 25mM $[Ru(dpp)_3]Cl_2$ in EtOH was pipetted into 6.82mL hydrolyzed sol. This doped sol was mixed on a touch mixer for 1 min. The mixed sol was stored in the dark in the refrigerator. Xerogel based sensor elements were formed by using 70µL of *sol* and spin-coated it on the surface of a microscope glass cover slip at 400 rpm for 30s giving the film a thickness of few microns.

A diode laser (λ=445nm) was used to excite the xerogel sensor placed in a test chamber. The $O_2$ concentration in the test chamber is controlled by a custom built flow-meter, consisting of a matched pair of air flow controllers connected to $O_2$ and $N_2$ gas cylinders. Given the lifetime detection range of the DTIM from 4µs to 14.4µs, we were only able to measure the lifetimes between 0% and 3% $O_2$ concentrations. The lifetimes measured were 4.95µs and 4.15 µs at 0% and 3% $O_2$ concentrations, respectively. The results approximately match the expected lifetimes [4]. In future, we aim to study the use of the DTIM system with luminophores that have longer decay lifetimes.

## IV. Conclusions

We developed and examined a CMOS IC for the direct measurement of luminescence lifetimes in miniaturized chemical sensors. The DTIM IC, fabricated in TSMC 0.35µm CMOS process, functioned to detect lifetimes from 4µs to 14.4µs and provides an 8-bit digital output. DTIM system can be tuned to detect longer lifetimes. The system allows compensation to overcome the response time delay of the photodetection and signal processing circuitry.

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