

Broad bandwidth air-coupled micromachined ultrasonic transducers for gas sensing

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1	BROAD BANDWIDTH AIR-COUPLED MICROMACHINED ULTRASONIC
2	TRANSDUCERS FOR GAS SENSING
3	
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26 Abstract

27 The present work aims to develop ultra-wide bandwidth air-coupled capacitive micromachined ultrasonic transducers (CMUTs) for binary gas mixture analysis. The 28 29 detection principle is based on time-of-flight (ToF) measurements, in order to monitor gas 30 ultrasound velocity variations. To do such measurements, CMUTs were especially designed 31 to work out of resonance, as a microphone. The chosen membrane size is $32x32 \ \mu m^2$ and gap 32 height 250 nm. The resonance frequency and collapse voltage were found at 8 MHz and 58 V 33 respectively. As mentioned, the CMUTs were exploited in quasi-static operating mode, in a 34 very low frequency band, from 1 MHz to 1.5 MHz frequency. The transducer impulse 35 response was characterised, and a -6 dB relative fractional frequency bandwidth (FBW) higher than 100 % was measured, enabling to use CMUT for the targeted application. 36 37 Additionally, a measuring cell has been designed to hold the fabricated CMUT emitter and 38 receiver prototypes facing each other. The volume inside the cell was kept lower than 3 mL 39 and the surface of emitter/receiver was 1.6×8 mm². To validate the general principle of the 40 proposed technique, two binary gas mixtures of CO_2/N_2 and H_2/N_2 , with varying 41 concentration, have been tested. The results are very promising with a measured limit of detection (LOD) of 0.3 % for CO₂ in N_2 and 0.15 % for H₂ in N_2 . 42

43 Index terms: Capacitive micromachined ultrasonic transducers, gas sensors, air-coupled
44 transducers, time-of-flight.

46 I. INTRODUCTION

47 Capacitive micromachined ultrasonic transducers (CMUTs) were introduced in mid 48 1990s by M.I. Haller and B.T. Khuri-Yakub [1], [2]. Since then, they are considered as a 49 potential substitute to the conventional piezoelectric transducers due to their numerous 50 advantages such as miniaturization, lead-free, relatively broad bandwidth and improved axial 51 resolution [3]. In the beginning, CMUTs were developed for airborne applications but, 52 rapidly, were used to address multiple applications, in particular medical ones [4]–[12]. As an 53 example, this leads to the development of dedicated CMUT probes for cortical bone 54 assessment [13] and dual-modality CMUT probes for imaging and therapy [14], [15]. The 55 performances of CMUT in air applications have been underestimated for long, due to their 56 lack of sensitivity for non-destructive testing applications [16]. Recently, new CMUT 57 structures were designed to produce ultrasound field in the frequency range of 50 kHz -58 150 kHz] [17], [18], showing encouraging performances with receiving sensitivity of 59 16.6 mV/Pa and fractional bandwidth up to 10 %. For the same frequency range, transducer topologies based on annular CMUTs was also proposed as an interesting approach to improve 60 61 the transmit efficiency [19]-[21], where value of 70 Pa/Volt was predicted. Air-coupled 62 CMUT transducer with a central frequency of 400 kHz was finally mentioned in the 63 literature [22]. A bandwidth of 1 MHz was claimed in the paper but experimental results 64 presented were not relevant enough to confirm such value. In the present work, the aim is to 65 develop a complete air-coupled CMUT transducer to serve as a binary gas sensor that works on the time-of-flight (ToF) measurement principle. The sensing principle consists of 66 67 measuring change in gas acoustic properties here, ultrasonic velocity, as already demonstrated 68 for monitoring of human-specific lung gas exchange [23] and for the wind speed 69 measurement [24]. In our case, the final application is the monitoring of hydrogen storage 70 sites [25], targeting to detect gas leaks, i.e. to measure the presence of low concentration of 71 hydrogen (typ. < 0.5 %).

The first research article on CMUT in the field of gas sensing was published in 2007 by Park *et al.* [26]. In their work, the top electrode was coated with a sensitive polymer in order to detect the molecule of interest, by increasing the vibrating plate mass and, thereby, decreasing the resonant frequency when absorbing these gas molecules. This paves the road to multiple researches that have been done and are ongoing on CMUT as a chemical/bio gas sensors [27]–[33]. However, adding sensitive coated polymer generally leads to only partial selectivity to multiple analytes. In addition, such solutions suffer from environmental effects, 79 which includes temperature or humidity variations but also material aging [34]. Therefore, 80 sensors without sensitive coating are preferable in particular when long-term stability is a 81 crucial issue and this even though their intrinsic selectivity limits applications [35]. In 82 previous work [36], [37], we demonstrated the relevance of using an uncoated CMUT 83 approach in order to sense binary gas mixture. In these works, the detection principle was 84 based on monitoring the electrical anti-resonance frequency shift, caused by gas density and 85 viscosity variations. Again, the targeted sensor was an uncoated CMUT transducer that detects ultrasonic velocity variations and, hence, gas compressibility and density variations. In 86 87 this paper, a clear objective is also to demonstrate the versatility of CMUT technology for such applications and to demonstrate their ability to detect variations in density, 88 89 compressibility as well as viscosity of gas medium.

90 It is worth to mention that the developed air-coupled CMUT transducer must meet 91 several specifications. First, in terms of global footprint, the fabricated sensor must be as 92 small as possible to be inserted in the dedicated measurement cell (few mL) developed 93 previously by our group [35]–[37]. Secondly, the ultrasonic gas velocity variations have to be 94 measured over a low travelling distance, typically few millimeters (< 10 mm), leading to very 95 low ToF variations of ultrasonic pulses to be detected (few µs). The detection of low 96 ultrasonic transit time variations is a quite current issue for various applications like ultrasonic 97 flowmeters [38], the blood flow velocity [39] or tissue motion detection [40]. Whatever the 98 application is, such ToF measurements make sense if i) the travel path corresponds to several 99 acoustic wavelengths and ii) the transducer impulse response duration is lower than emitter -100 receiver transit time. This means that both working frequency and fractional frequency 101 bandwidth (FBW) have to be high enough to meet these operating conditions. To achieve 102 such conditions, the CMUT used in this study will have working frequency ranging between 103 1 MHz and 1.5 MHz with a FBW higher of 50%.

104 Several air-coupled transduction technologies exist in the literature; from devices 105 based on bulk piezoelectric plate [41] to electrostatic transducers based on Mylar film 106 vibration [42]. Recently, based on piezoelectric micromachined ultrasonic transducer (PMUT) 107 technology, the first millimeter accurate ultra-low power ToF sensor has been launched 108 commercially, used for proximity sensing applications with a dimension of 15 mm² [43], [44]. 109 Few other PMUT applications as microphones, using ToF technique, can also be found 110 in [45], [46]. Finally, a novel 2D flexural ultrasonic transducer array, working on ToF 111 principle and composed of eight piezo ceramic disc elements with a total dimension of 112 36x36x0.25 mm³, has been demonstrated for accurate average flow velocity [47]. One can

113 note that, for this work, the travelling distance was ranging from centimeter to meter scale 114 [48], meaning the FBW of fabricated devices was not a crucial issue for the targeted 115 applications. Typically, FBW of 20 % are enough for such working distance range. To 116 achieve FBW values higher than 50 %, the developed air-coupled CMUTs were aimed to 117 work like microphone, not only in reception, as shown in references [49]–[51], but also in 118 emission mode. This design strategy is clearly novel compared with previous works where the 119 membrane dimensions (sizes and thicknesses) were optimized to reach resonance frequencies 120 close to the targeted working frequency [17]–[21], [52], [53]. To achieve such low resonance 121 frequencies, the mechanical stiffness of the membrane must be adjusted, either by changing 122 the CMUT sizes or thicknesses. However, if the diaphragm rests on a vacuum cavity, it 123 undergoes significant mechanical deformation due to atmospheric pressure, which can cause 124 non-functionality of CMUT (i.e. collapsed cells). There are two possible solutions to get 125 circumventing such constraint. The first is to perforate the rear side [17], [18] of the cavity in 126 order not to be affected by atmospheric pressure. The second is to increase the cavity height 127 (typically up to 10 µm thick) [20], [53] to ensure that the membrane could deflect without 128 collapsing. Therefore, the CMUT fabrication process for air applications requires specific 129 steps that differ from those used for immersion applications. In our case, we have deliberately 130 chosen to keep the CMUT diaphragm on a vacuum cavity (i.e. same process for immersion 131 applications) and to exploit CMUT at working frequencies far from the resonance frequency, 132 in a quasi-static operating mode as used for microphones. The CMUT were designed in order 133 to keep membrane deformation amplitude as low as possible on one hand and to increase the 134 radiated acoustic power on the other hand, using frequency of interest in the range of 1 MHz 135 to 1.5 MHz. In former work [54], we have shown the relevance of such approach without 136 evidencing design strategy, precise gas concentrations records and without discussion on 137 acoustic velocity measurements. In this study, the gas concentration is monitored accurately 138 for two different gas mixtures and compares the obtained results with theoretical predictions. 139 Further, the limit of detection (LOD) and sensitivity of CMUT gas sensor will be presented in 140 § IV.B.

The presented paper consists of three main sections. Section II gives an overview of the CMUTs fabrication process, CMUTs design, CMUTs packaging and the description of the measuring cell. From design considerations, one prototype was fabricated and characterised. Results are described in section III, using both electrical impedance data and electroacoustic measurements. Next, section IV presents the test of the fabricated gas sensor. To demonstrate the sensor efficiency, two gas mixtures were investigated, first carbon dioxide (CO₂) in 147 nitrogen (N₂) and then, hydrogen (H₂) in N₂. This work shows that the developed device 148 enables to detect very low variations in the transit time for very low gas concentration (e.g. <149 1 %).

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152 II. DESIGN, FABRICATION, PACKAGING OF CMUT AND MEASURING153 CELL

154 A. CMUT fabrication process-flow

The silicon nitride (SiN_x) based CMUT used here was initially developed for medical imaging and therapeutic purposes [13], [55]. Although the CMUTs were manufactured using standard surface micromachining techniques [56], our group improved the process over years by distinguished choice of materials and deposition techniques [7], [9], [42], [57]. A brief process flow is illustrated in Figure 1.

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Figure 1: CMUT fabrication process flow; (a) deposition of insulation layer (SiO₂), bottom electrode (PolySi) and sacrificial layer (PSG); (b) patterning of bottom electrode and sacrificial layer; (c) deposition of membrane layer (SiN_x); (d) removal the sacrificial layer and releasing the membrane; (e) sealing the membrane; (f) deposition of top electrode (Al).

In step 1 (Figure 1.a), a 6-inch Si (100) wafer with high resistivity is used. To ensure electrical insulation of the Si substrate, a 1.2 μm thick silicon dioxide layer has been grown by thermal oxidation. Then, for bottom electrode, a 450 nm thick polycrystalline silicon (PolySi) layer has been deposited on the oxide layer using Low Pressure Chemical Vapour Deposition (LPCVD). The PolySi layer was heavily doped with POCl3 in order to ensure

172 high electrical conductivity. The second step consists in a crucial sacrificial layer 173 (PhosphoSilicate Glass oxide, PSG) deposition, as its thickness defines the cavity height of 174 the CMUT device. The step 2 (Figure 1.b) consists in a two processes: the PSG dry etching to 175 define the membrane geometry and another etch step to define the bottom electrode (PolySi). 176 In Step 3 (Figure 1.c), the CMUT membrane is deposited by LPCVD process, hereby a SiN_x 177 material with thickness of 400 nm. Step 4 (Figure 1.d) is a chemical etching using hydrofluoric acid to remove the sacrificial layer and to release the membrane. Step 5 178 179 (Figure 1.e) consists in the cavity sealing and finally step 6 (Figure 1.f) correspond to 180 aluminium top electrode deposition and patterning (450 nm). Later, the fabricated CMUTs are 181 diced and ready for characterisation and packaging.

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B. CMUT and transducer design

184 CMUTs were designed in such a way that they are able to work in a low frequency 185 band, far from their own resonance frequency. In such case, they behave like a microphone 186 and enable to obtain the broadest frequency bandwidth and lowest impulse response duration, 187 as possible. From the previous studies, it was shown that, when CMUTs work in quasi-static 188 regime, non-linear time-domain models enable to predict accurately the transducer 189 response [58], [59]. The major design criterium was based on the output acoustic power 190 radiated by the transducer. For a given 2D array of CMUTs, the output acoustic intensity (the 191 mean spatial acoustic power) P_a is written as:

$$P_a = \frac{1}{2} R_a \left(2\pi f_0 \frac{u_0}{3} \right)^2$$
 Eq. 1

192 Where R_a , is the real part of the transducer radiation impedance, f_0 is the ultrasonic wave 193 frequency and u_0 is the maximum peak displacement amplitude of the CMUT, taken at its center. Here, it is assumed that two-dimensional periodic boundary conditions can be applied 194 195 to each CMUT, R_a , for such boundary conditions, equals to the acoustic medium impedance 196 multiplied by the surface coverage ratio of the transducer. To consider the fact that the 197 membrane displacement is non-uniform, a factor 1/3 was applied to obtain an approximate 198 value of the spatial mean membrane displacement. This value was obtained through time-199 domain simulations, in which all details are given in reference [59].

To approximate u_0 , it has been considered that, when CMUT works like a microphone, the membrane is able to sweep the whole gap height, from the static membrane position (U_i), taken at the center, up to the bottom electrode.

$$u_0 = \frac{(h_{gap} - U_i)}{2}$$
 Eq. 2

204 Where, h_{gap} is the total gap height.

By considering Eq. 1 along with the design parameters obtained from the process specifications given in Table 1, P_a was computed for square CMUT membranes, along with the spatial distribution presented in Figure 2 for dimension 'w' ranging from 10x10 μ m² to 80x80 μ m².

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Table 1: CMUT material parameters and thicknesses

Layer	SiN _x	Al
Thickness (nm)	400	450
E (GPa)	220	68
v (poisson ratio)	0.22	0.35
ρ (kg/m3)	3300	2700
ε _r	7.5	





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Figure 2: Sketch of CMUT distribution over the surface of the transducer, w is the width of each CMUT, d_x the CMUT-to-CMUT distance along X axis, and d_y along the Y axis. Black squares represent the front electrodes, the surface area of which corresponds to 50 % of the CMUT surface area

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218 The active surface ratio was given by the parameter $w^2/(w+d_x)(w+d_y)$ as presented in Figure

219 2. d_x and d_y values were fixed by process specifications. More precisely, d_x is higher than d_y

because holes used to release CMUT membrane were placed along X axis between two CMUTs. For each membrane size, the initial deflection, caused by the atmospheric pressure, was computed with finite difference model described in [60], and the h_{gap} value was fixed to 250 nm. The computed output acoustic intensity according to the membrane size is shown in Figure 3, for two frequencies in the range of interest, i.e. 1 MHz and 1.5 MHz.



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Figure 3: Computed output acoustic intensity radiated in air for square CMUTs with 2D
 periodic layout and membrane size ranging from 10 μm to 80 μm

228

Figure 3 clearly highlights the existence of an optimal size range where the acoustic intensity reaches a maximum, here it is between 30 μ m and 40 μ m width. This maximum comes from a trade-off between low size CMUT for which the initial deflection caused by atmospheric pressure is low and large size CMUT for which the active surface ratio is high. Hence, the final geometry retained was 32x32 μ m². All geometrical specifications are reported in Table 2. The presented collapse voltage and resonance frequency are obtained experimentally from electrical impedance measurements and will be discussed in § III.A.

236

For these first experiments, since no prior knowledge on radiation surface was identified to have a suitable signal-to-noise ratio, it was targeted to fabricate an array of single element transducers for which the surface can be adjusted. Therefore, a multi-element array of eight independent acoustic emitters was fabricated. Each element was rectangle with 8 mm length and 0.8 mm width, which corresponds to 2.4 acoustic wavelength in air for 1 MHz working frequency and 3.6 wavelength at 1.5 MHz. The final number of emitters used (from 1 to 8) for

- the experiments was defined from preliminary experiments presented in [54]. It was observed
- that pairing elements was enough for the targeted working distance, i.e. less than 10 mm.
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Table 2: Specifications of the fabricated transducer

CMUT size	32x32 µm²
Gap height	250 nm
Area of top electrode	20x20 µm²
Active surface ratio	40 %
Collapse voltage (V _c)	58 V
Resonance frequency	8 MHz
Transducer length	8 mm
Element width	0.8 mm
Number of elements	8

249 C. CMUT packaging and measuring cell description

250 To package the CMUT array, a specific printed circuit board (PCB) was designed and 251 fabricated as shown in Figure 4. The diced eight element array of size 6.4x8 mm² was glued 252 on PCB (refer Figure 4.a) using an insulating epoxy adhesive (ALBEBOND 84-3J by Henkel) 253 and kept in a lab oven for annealing during one hour at 150 °C. Then, wire bonding was done 254 using K&S 4532AD manual wedge bonder with fine aluminium wedge bonding wires of 255 25 µm diameter. Epoxy NoSWEEP from Polysciences (wire bond encapsulant) was used to 256 protect the bonded wires. The eight elements array was paired together to form four acoustic 257 emitters/receivers. Then, the prototype was annealed at 100 °C and 165 °C for one hour each, 258 to harden the encapsulation. Lastly, to complete the emitters/receivers prototype packaging, 259 UFL connectors were soldered on the rear face of the PCB for electrical connections.

260

A specific measuring cell was designed as a gas sensor unit. The cell was made of acrylic monomer and fabricated using 3D printing with cubic shape. It includes two large openings, one in the front and other in the rear face (18 mm in diameter, see Figure 4.b) to conform the emission and the reception transducers. Both PCBs were positioned face-to-face to perform ToF measurements. The chamber sealing was ensured by gaskets, which were placed between the PCBs and each side of the acrylic cell to prevent gas leakage. Two small openings on the top and bottom side of the cell were kept for gas inlet and outlet respectively (see Figure 4.c). The measuring cell was fabricated in such a way that the propagationdirection of ultrasound is always perpendicular to the gas outflow.

270

To master precisely the concentration of binary gas mixtures, the measuring cell geometry such as height (h) (distance between emitter and receiver) and volume (V) of the inner cylinder were chosen with great care. The total volume of the inner cylinder was kept less than 3 mL in order that the filling time of the cavity be small enough to apply in real time gas concentration command. The distance between the emitter and the receiver was fixed accordingly, at a value close to 7 mm.

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Figure 4: a) emitter/receiver prototype b) inner view of the measuring cell c) completely packed gas unit for ToF measurement

282 III. CMUT CHARACTERISATION RESULTS

A. Electrical impedance measurement

284 Electrical impedance measurement of developed CMUT devices makes it possible to 285 extract most of the information about their electrical and mechanical behaviour. The 286 impedance measurement protocol presented here is the same as the one shown in 287 reference [60]. HP4294A impedance analyser, from Keysight Technology, combined with 288 4294A1 connected probe was used. The CMUT biasing voltage was swept automatically 289 (with a MOTECH DC supply) from 0 to 90 V with a pitch of 2 V. For each biasing value, the 290 electrical impedance was acquired from 1 MHz to 15 MHz, in the frequency band where 291 mechanical resonance occurs. With the obtained data, most of the electromechanical 292 parameters can be derived such as, capacitance at low frequency (measured at 1 MHz), 293 resonance frequency (f_r) , antiresonance frequency (f_a) and electromechanical coupling 294 coefficient (k_t) . k_t was assessed through the following approximate relationship:

$$k_t = \sqrt{\frac{f_a^2 - f_r^2}{f_a^2}}$$
Eq. 3

The collapse voltage value, V_c , was determined as the voltage from which the capacitance variations suddenly drops, meaning that a part of the CMUT membrane is stuck to the bottom cavity. An experimental value of 58 V was found for the 32x32 μ m² cells. Note that more than ten prototypes were realized and the collapse voltage was the same for each.

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Electrical impedance measurements for three biasing voltages are plotted in Figure 5, with a focus around the resonance frequency. CMUT works as expected, i.e. the electrical antiresonance frequency decreases on increasing the biasing voltage. The mechanical resonance frequency can be extracted at very low biasing voltage value; one can guess a value close to 8 MHz. Figure 6 demonstrates the electromechanical coupling coefficient, which increases by increasing the bias voltage and reaches a maximum of 60 %, just before collapse event.







Figure 5 : Electrical impedance measurement results of single element CMUT in air



Figure 6 : Experimental electromechanical coupling coefficient of CMUT according to the
 biasing voltage

B. Electroacoustic characterization in air

314 The ultrasound ToF measurement was initially performed in air before testing in gas 315 medium, to check several parameters such as maximum distance between emitter and 316 receiver, emission frequency (f_e) , as well as the quality of the received signals in terms of 317 signal-to-noise ratio. For the operating excitation conditions, biasing voltage value was fixed 318 at 70 % of V_c (i.e. 40 V) and the excitation voltage was one period of sinus-pulse with an 319 amplitude of 30 % of its V_c (amplitude of 15 V). Five working frequencies were tested 320 500 kHz, 1 MHz, 1.5 MHz, 2 MHz and 3 MHz. Emitter and receiver were placed face-to-face 321 with a separation distance of 7 mm. To provide the AC excitation signal, a home-made 322 analogical emitter was used, similar to the one used for the therapy-imaging platform 323 described in reference [14]. Charge preamplifier circuit from Cooknell electronics ltd. (CA7/c 324 model with 250 mV/pC gain) was used to drive the electrical signal delivered by the receiver. 325 Signals were acquired with Picoscope 2000 series acquisition card (12 bits resolution and 326 20 MHz sampling frequency) placed on the receiving end. The time domain and frequency 327 domain response of the transmitted signals are plotted in Figure 7 and Figure 8, no average 328 was applied to the signals. Principally, CMUT is emphasized as a microphone based on its 329 design, which works far away from its resonance frequency band. For the five excitation 330 frequencies, the received signal has significant voltage amplitude, with a spectrum clearly 331 shifted to higher frequencies as the excitation frequency increases.





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Moreover, the -6 dB relative FBW of the received signals clearly follows the ones of the excitation signal. For example, at 1 MHz, 105 % FBW was measured. At 500 kHz and 1 MHz, a spectral contribution arising from non-linear intrinsic behaviour of CMUT has been observed, similarly at 1.5 MHz respectively. For 2 MHz and 3 MHz excitation frequencies, there is a gain in high frequency components, but attenuation in air compensated for this frequency shifting meaning that the frequency where the maximum occurs remain at 1.5 MHz.





Figure 8 : Frequency domain response of the transmitted signal for different excitation frequencies

348 349 The second step of this characterisation was to assess the maximum working distance between 350 emitter and receiver, not for the application targeted in this study, but for potential future 351 applications. The experiments were done at 1 MHz excitation frequency. The emitter was kept

352 at fixed position while the receiver has moved along the emitter axis from 5 mm to 250 mm. 353 Data acquisition conditions were kept similar as of the previous experiments. The set of 354 impulse responses received are gathered and plotted in Figure 9. The corresponding frequency 355 domain responses for 5 mm, 100 mm and 220 mm are plotted in Figure 10. With the obtained 356 data, it is clear that the Limit Of Detection (LOD) in terms of distance for this device is 357 220 mm maximum, beyond that the signal-to-noise ratio was not high enough. The received 358 signal shows 30 dB of dynamic with the pulse shape that is clearly centered at 1 MHz and 359 shifts toward lower frequencies when the distance increases, due to acoustic attenuation in air. 360 Although these CMUTs were not designed to attain the optimized sensitivity at larger 361 distances but with the frequency bandwidth, the maximum working distance experiment 362 performed using pulse-echo mode paves the way for other applications. Furthermore, with the 363 obtained data, it is clear that this setup can be used for imaging applications in air, based on 364 its exceptional performances in terms of lateral and axial resolution. In addition, works are currently under investigation to perform imaging tests based on the Synthetic Aperture 365 366 Focusing Technic beamforming strategy [17], [61], [62].



Figure 9: Time-domain signals received for a travelling distance ranging from 5 mm to 220 mm at the excitation frequency of 1 MHz

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Frequency (MHz)
 Figure 10: Frequency response of the received signal for 5 mm, 10 mm and 220 mm working
 distances between emitter and receiver, excitation frequency of 1 MHz

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IV.

A. Time-of-flight setup

TIME-OF-FLIGHT IN GAS MEDIUM

The fabricated gas sensor unit was tested using a specific test-bench dedicated to the characterization of binary gas mixtures as shown in Figure 11. For the experiments presented in this paper, two binary mixtures were tested: carbon dioxide (CO₂) or hydrogen (H₂) in nitrogen (N₂). Nitrogen was chosen as the reference gas because it has an acoustic velocity close to that of air at room temperature. The test-bench therefore includes three gas tanks (CO₂, H₂ and N₂) and electro-valves controlled by a Labview programme allowing binary mixtures (CO₂/N₂ or H₂/N₂) to be made with chosen concentrations.





387

Figure 11: Diagram of the test-bench used for the characterization of binary gas mixtures

389 As explained in the introduction, the sensor is based on the measurement of time of flight 390 (ToF) variations that depend on the speed of sound in the gas mixture. To give an order of 391 magnitude of the time variations to be measured, the values of the ToF for an acoustic wave 392 to travel a distance of 7 mm in the three pure gases (CO₂, H₂ and N₂) are given in Table 3. It 393 should be noted that the ToF difference between pure CO_2 and pure N_2 is only 6 µs, which 394 means that it is necessary to be able to measure very small ToF variations to accurately 395 monitor the concentration of the binary mixture.

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- 397

Table 3: Physical properties of CO₂, N₂ and H₂ gases [63]–[65]

Gas	Density ρ (kg/m ³)	Isobar Specific Heat c _p (J/°C.mol)	Isochoric Specific Heat c _v (J/°C.mol)	Velocity C (m/s)	Expected time of flight for pure gas (μ s) at d \approx 7 mm
CO_2	1.82	37.1	28.8	267	26
N_2	1.16	29.1	20.8	349	20
H_2	0.083	28.9	20.5	1306	5.4

398

399 To check the initial working conditions, a first measurement was done by filling the cell with 400 pure N₂ and then with pure CO₂. To carry out this experiment, the overall voltage was kept at 401 90% of V_c with the bias $V_{DC}=32$ V and the alternative component $V_{AC}=20.5$ V (41 V peak-to-402 peak). The frequency is 1.5 MHz, as it corresponds to maximum of the spectrum amplitude 403 (see Figure 8). The bandwidth has been deliberately reduced to avoid energy transmission in 404 the frequency band that is not used in the reception mode (attenuation losses). The emitted 405 signal has therefore been modified: it is a sine burst of 1.5 period instead of 1 period.

406

407 The data acquisition duration has been chosen longer than previously to be able to see the 408 forward contribution from emitter to receiver (Figure 12). Note that the receiver signals for 409 both N_2 and CO_2 are clearly separated from the emission, meaning that the duration of the 410 transducer impulse response is small enough to accurately monitor the ToF variations.

411

412 The reception occurs after a delay τ which depends on the gas since a difference between pure 413 N_2 and pure CO_2 is clearly observed, with a value close to the expected one of 6 µs. For intermediate CO₂ concentration values, it was chosen to detect the transit time between 414 415 emission and reception. A standard method based on the detection of the position of the 416 minimum of the signal was used to identify the ToF shift. Since the sampling frequency was 417 high enough (20 MHz), it was possible to interpolate data [39] between two sampling points, 418 resulting in a noise standard deviation of 6 ns instead of 20 ns without interpolation. As 419 expected, the shape of the received signal is slightly different from the results shown in 420 Figure 7 since the excitation signal was longer.





423 Figure 12 : Alternative part of the reception signal ($V_{DC} = 32 \text{ V}$, $V_{AC} = 20.5 \text{ V}$) under pure N_2 424 (solid line) and pure CO_2 (dotted line)



426 As a first proof of concept, tests were carried out for the binary mixture (CO_2/N_2) with 427 concentration steps from 20 % to 100 %. Between each step, pure N_2 is sent to the measuring 428 cell to show the reversibility of the sensor. Using a set of mass-flow controllers (Brooks 429 5850S), the gas flow is kept constant at 100 ± 0.7 cm³.min⁻¹ with a stabilization time of about 45s. The temperature T is kept at 20 ± 2 °C. The results (Figure 13.a) clearly show that the 430 431 proposed setup using two CMUTs to measure ToF allows a change in the gas composition to 432 be detected. The average relative ToF values as a function of the concentration steps are 433 shown in Figure 13.b. In order to confirm the correlation between the sensor output and the 434 gas concentration, the relative changes in ToF have been determined theoretically 435 (Figure 13.b).

436

437 Although a detailed demonstration (refer Appendix) is out of this scope, it can be shown that 438 the relative change in the ToF of a gas with respect to N_2 is given by:

$$439 \qquad \frac{d\tau}{\tau} = 1 - \frac{C}{C_{N2}}, \qquad \text{Eq. 4}$$

440 where C_{N2} and *C* are the sound velocity under pure N₂ and binary mixture, respectively. *C* 441 depends on the molar fraction of the targeted gas (CO₂ or H₂) x_G according to:

442
$$C = \sqrt{\frac{x_G c_{p,G} + (1 - x_G) c_{p,N2}}{x_G c_{v,G} + (1 - x_G) c_{v,N2}}} \frac{P_0}{x_G \rho_G + (1 - x_G) \rho_{N2}}$$
Eq. 5

443 where ρ , c_p and c_v are mass density, the isobar heat capacity and the isochoric heat capacity, 444 respectively. For all properties, the subscripts *G* and *N*₂ designate the targeted gas (CO₂ or H₂) 445 and N₂, respectively. All these parameters are given in Table 3.

446 In the same way (full demonstration in the Appendix), for x_G close to zero the sensitivity S_0 is 447 given by:

448
$$S_0 = \frac{1}{x_G} \cdot \frac{d\tau}{\tau} = \frac{1}{2} \left(\frac{\rho_G}{\rho_{N2}} + \frac{c_{\nu,G}}{c_{\nu,N2}} - \frac{c_{p,G}}{c_{p,N2}} - 1 \right)$$
 Eq. 6

The good agreement between theory and experiment confirms that the variations of the output are mainly due to changes in gas concentration. A slight deviation can be noted which results in a smaller sensitivity than the theoretical one. There are two possible reasons for this small 452 discrepancy. The first is due to the method used to monitor the ToF variations, i.e. the method 453 based on detecting the minimum peak position of the received pulse. Although this aspect is 454 out of the scope of this paper, a potential solution would be to make the detection system 455 more robust by implementing, for example, a cross-correlation detection. The second possible 456 reason for the discrepancy between theory and experiment is the initial ToF value, 20 µs was 457 chosen. In practice; it is difficult to obtain an exact value of the initial ToF because the 458 distance between the emitter and the receiver as well as the initial ultrasonic velocity must be 459 known acurately. So, obtaining an absolute value of the initial time-of-flight requires an 460 extremely complex protocol, using a second reference signal. The implementation of such 461 method is clearly out of scope for this investigation. Practically, implementing a calibration 462 procedure, from which the sensitivity parameter is determined, is preferable.



463

Figure 13 : Measured relative change in τ for different values of x_{CO2} (100%, 80%, 60%, 40% and 20%). (a) As a function of time ($V_{DC} = 32V$, $V_{AC} = 20.5V$). (b) As a function of x_{CO2} (dots) with theoretical values of Eq. 5 (solid line) and the tangent at the origin from Eq. 6 (dashed line)

468

In order to characterise the proposed method for lower concentration, mixtures of either H₂ or CO₂ in N₂ were tested at different low concentration values (4%, 1%, 2%, 3% and 4%). The results are shown in Figure 14. A drift that is too small to be noticed for high concentrations can now be appreciated and is consistent with temperature variations *dT* below 1°C according to Eq. 7 (refer Appendix for demonstration). After removing such drift by a simple polynomial fitting, the calibration curves were obtained and are shown in Figure 15.

$$475 \qquad \frac{d\tau}{\tau} = -\frac{1}{2}\frac{dT}{T}$$
 Eq. 7

These results highlight a key feature for the sensor: its output varies linearly with the physical quantity to be detected, as long as the relative variations are only a few percent. Despite the 478 simplicity of the model (ideal gas, small concentrations) and detection method, the sensitivity

479 values (magnitude and sign) remain consistent with the ones obtained by using the Eq. 5 480 $(3.4 \times 10^{-3})\%$ for CO₂ and -7.1 $\times 10^{-3}\%$ for H₂).



481

482 Figure 14: Measured relative change in τ by alternating pure N₂ and either CO₂ (a) or H₂ 483 (b) at different concentrations (4% 1% 2% 3% and 4%)



484

485 Figure 15: Sensor output characteristics for CO₂ (dashed line) and H₂ (solid line)

486



$$488 \qquad \text{LOD} = \frac{3\sigma}{S_0} \qquad \text{Eq. 8}$$

Where $\sigma = 29 \times 10^{-5}$ is the noise standard deviation of the measurement of $d\tau/\tau$. Since the LOD depends on *So*, its value depends on the targeted gas. The estimated values for H₂ and CO₂ are approximately 0.15% and 0.3%, respectively. In order to verify them experimentally, similar experiments as the ones of Figure 14 were performed with the appropriate concentrations and the results are presented in Figure 16.



496Figure 16: Measured relative change in τ (drift removed) by alternating pure N2 and either497CO2 (a) at 1 %, 0.5 %, 0.3 %, 0.1 % and 1 % or H2 (b) at 0.5 %, 0.3 %, 0.15 %, 0.05 % and4980.5 %. These concentrations were chosen to estimate the LOD for CO2 and H2499

500 These measurements validate the calculated values for the LOD. A summary of the 501 experimental sensor performance parameters is presented in Table 4.

Table 4: Summary of the experimental sensor performance

Gas	S ₀	LOD
Gas	(10 ⁻³ /%)	(%)
CO ₂	2.85	0.3
H ₂	-4.9	0.15

514 V. CONCLUSION AND PERSPECTIVE

515 A new approach for binary gas mixture monitoring using uncoated sensors was 516 investigated and demonstrated. The developed method is based on the gas acoustic properties 517 measurement, i.e. the ultrasound velocity, combining usual ToF detection principle and ultra-518 wide bandwidth air-coupled capacitive micromachined ultrasonic transducers. Air-coupled 519 CMUT were designed as microphone in order to reach FBW higher than 50 %. Experimental 520 results, based on fabricated prototypes, demonstrated the approach relevancy since FBW 521 higher than 100 % were obtained, with a tunable central frequency, from 500 kHz up to 522 2 MHz. The performance in terms of signal-to-noise ratio showed that, with the fabricated 523 prototype, a working distance of a few centimeters is possible. This paves the way for new 524 applications such as acoustic imaging in air.

525 The ToF measurements performed with these first prototypes have proven that this 526 technique is suitable to monitor accurately binary gas mixtures. In this paper, CO₂ in N₂ and 527 H₂ in N₂ were tested but it can be widely used for other gas mixtures. Sensor performance was 528 evaluating by measuring both the sensitivity at low gas concentration values and the limit of 529 detection (LOD). For both gas mixtures, the experimental sensitivity is generally in agreement 530 with the theoretical value despite deviations that could possibly be reduced by improving the detection algorithm. In the case of H2 detection, concentration lower than 0.15 % have been 531 532 detected.

It is certain that this LOD value is not competitive with that of the state of the art of coated sensors. However, the approach presented in this paper shows a good trade-off to meet simultaneously good accuracy, simplified fabrication method, high integrability level and high reliability. This homemade setup is portable. The sensor performance can be improved in two ways: i) using optimized electronic driving circuits in reception, ii) using a wafer bonding process, since it enhances the homogeneity of CMUTs and the active surface ratio too.

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546 VI. ANNEX : theoretical determination of the relative time-of-flight variations

547

548 The Time of Flight τ depends on both the sound velocity of the gas *C* and the distance *d* 549 between the emitter and the receiver:

550
$$\tau = \frac{d}{C}$$
 Eq. A.1

551

The relationship between the gas properties and the sound velocity *C* is given by Newton-Laplace formula [67]:

554
$$C = \sqrt{\frac{\gamma P_0}{\rho}}$$
 Eq. A.2

where P_0 is the static pressure, ρ the mixture mass density and γ the heat capacity ratio. In the case of binary gas mixture, both properties, ρ and γ , depend on the molar fraction of the gas to detect x_G . The mixture gas density in N₂ is given by:

558
$$\rho = x_G \rho_G + (1 - x_G) \rho_{N2}$$
 Eq. A.3

so where ρ_G and ρ_{N2} are the mass density of the pure gas to detect and pure N₂, respectively.

560 Additionally, the heat capacity ratio is given by Eq. A.4 [68]:

561
$$\gamma = \frac{x_G c_{p,G} + (1 - x_G) c_{p,N2}}{x_G c_{v,G} + (1 - x_G) c_{v,N2}}$$
 Eq. A.4

562 Where, $c_{p,G}$, $c_{p,N2}$ are the isobar heat capacity of the pure gas to detect and pure N₂, 563 respectively, $c_{v,G}$ and $c_{v,N2}$ are the isochoric heat capacity of the pure gas to detect and pure 564 N₂, respectively.

565

566 By combining Equations A.1 to A.4 it is possible to obtain a equation of the relative change in 567 time of flight, $d\tau/\tau$, when the initial state is pure N₂ (Eq. A.5):

568
$$\frac{d\tau}{\tau} = \frac{1}{2} \left(\frac{\rho_G - \rho_{N2}}{x_G \rho_G + (1 - x_G) \rho_{N2}} + \frac{c_{\nu,G} - c_{\nu,N2}}{x_G c_{\nu,G} + (1 - x_G) c_{\nu,N2}} - \frac{c_{p,G} - c_{p,N2}}{x_G c_{p,G} + (1 - x_G) c_{p,N2}} \right) x_G \quad \text{Eq. A.5}$$

569 which can be further simplified, in the case $x_G \ll 1$ into:

570
$$\frac{d\tau}{\tau} = \frac{1}{2} \left(\frac{\rho_G}{\rho_{N2}} + \frac{c_{\nu,G}}{c_{\nu,N2}} - \frac{c_{p,G}}{c_{p,N2}} - 1 \right) x_G$$
 Eq. A.6

571 From Eq. A.6, one can define the sensitivity for small concentrations S_0 (x_G as input and $d\tau/\tau$ 572 as output):

573
$$S_0 = \frac{d\tau}{\tau x_G} = \frac{1}{2} \left(\frac{\rho_G}{\rho_{N2}} + \frac{c_{\nu,G}}{c_{\nu,N2}} - \frac{c_{p,G}}{c_{p,N2}} - 1 \right)$$
Eq. A.7

574 Defined in such a way, S_0 depends only on the thermodynamic properties of the gases 575 involved and it is independent on the setup.

577 For an ideal gas, the mass density depends on the static pressure and temperature:

578
$$\rho = \frac{P_0 M}{RT}$$
 Eq. A.8

579 Then, assuming that γ is independent of the temperature, the dependence of the ToF to the 580 temperature can be expressed:

581
$$\frac{d\tau}{\tau} = -\frac{dC}{C} = -\frac{1}{2} \left(\frac{d\gamma}{\gamma} + \frac{dP_0}{P_0} - \frac{d\rho}{\rho} \right) = -\frac{1}{2} \left(\frac{d\gamma}{\gamma} + \frac{dT}{T} \right) - \frac{1}{2} \frac{dT}{T}$$
Eq. A.9

593	AUTHOR CONTRIBUTIONS
594	
595	PS and LI has equally contributed in the paper writing. PS, JFM, DA, LC and DC have
596	designed and fabricated the CMUT and the measuring cell. Further PS and DC has optimized
597	the parameters and made characterisation in air. LI and ID has validated the system by
598	performing ToF modelling and measurement with various binary gas mixtures. All the authors
599	have read and approved the manuscript.
600	
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