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Response Time Improvement of Gas Density and Gas Composition Sensor

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Abstract: We report on response time investigations of a commercial industrial gas density sensor. The sensor is typically applied to both pure gases and gas mixtures and can be used as stand-alone sensor or as part of a sensor system for gas composition determination of binary gas mixtures. The core element of the density sensor, a micro-mechanical quartz oscillator, is typically protected from dust and corrosive contaminant gases by a standard built-in filter package. This filter package determines the diffusion rate of the gas to the quartz and thus the response time. In order to relate this gas dynamics to the filter type, we tested sensors of different filter configurations (thickness and pore size) for pure gas (nitrogen, sulfur hexafluoride) and for gas mixtures (nitrogen plus perfluoroketone) and measured the respective response time. We found that changing or removing a specific element of the filter package can improve the response time by one to two orders of magnitude. These findings will trigger a novel sensor package for applications where fast sensor response is critical.

Keywords: Gas density, Quartz resonator, Sensor package, Filter, Gas diffusion, Teflon membrane, Sensor response time, Binary gas mixture, Gas composition.

1. Introduction

In various industrial areas, the composition of a specific gas component in a mixture needs to be known for efficient, reliable and safe operation. Here, we target the concentration determination of the various components in a novel electrical insulation gas [1]. For our and many other applications, laboratory analysis of the gas, e.g., by gas chromatography or spectroscopy, would be disadvantageous, since this is too complex and time consuming. Alternatives to these high-end solutions are, for example, systems based on acoustic sensing principles, on thermal measurements, or on damping effects of resonant mechanical oscillators. The sensor system reported here is based on the measurement of pressure, temperature, and density, whereas the density is determined using a resonant mechanical oscillator. The knowledge of the gas parameters (p-T-ρ) allows the determination of the gas composition of a binary gas mixture [2].

Besides the novel application of the density sensor to gas mixtures, also the conventional application to pure gases, such as sulfur hexafluoride (SF_6) is very important for us. Sulfur hexafluoride is used in gas-insulated switchgear because of its outstanding

insulation and arc quenching properties. It thus makes it possible to set up complex and safe power distribution systems even in confined spaces. The insulation strength depends basically on the gas density. The safety of the facility is guaranteed when the correct level of gas density is maintained within the enclosed systems. The tightness of the installation has to be checked and controlled as a leakage would result in a decrease in the insulation properties. Gas density, as an indicating parameter of potential leakage, is often indirectly determined by the gas pressure using manometers or pressure sensors. As the pressure in the hermetically closed volume varies enormously with temperature, such devices need temperature compensation, which are a source of errors. Trafag gas monitoring devices measure the gas density directly with the unique gas density reference principle or the patented quartz tuning fork technology, developed at ABB Corporate Research.

In this paper, we report on response time investigations of gas density sensors, we identified the critical sensor package element that limits fast response and suggest a novel sensor package leading to a significant response time improvement of one to two orders of magnitude.

2. Package of Density Sensor

For the application of gas composition measurement in binary gas mixtures, the quartz element in the density sensor [3] represents a key element in the (p-T-ρ) sensor system, for which highly accurate density readings are required (to a precision of $\Delta\rho \approx 5$ g/m^3). In order to reach and maintain the accuracy over time, filters are used to protect the quartz and thus to avoid degradation due to contact with particles and/or corrosive gas. A molecular sieve (zeolite filter) is used to absorb corrosive gas (e.g., HF). A metal sinter filter and a Teflon membrane are used against fine particles. The metal sinter filter is a disk of a thickness of 3 mm and a diameter of 6 mm and a pore size of 500 µm – 850 µm. The standard filter package of the density sensor is shown in Fig. 1. Fig. 1A shows a photo of the density sensor (type 8874) of which a sectional sketch with standard filter package is shown in Fig. 1B and Fig. 2 (left). The standard filter package consists of two metal sinter filters (14), a zeolite filter (19), and a Teflon filter membrane (16a) with pore size of 1 µm (diameter: 6 mm and thickness: 0.26 mm).

In our previous studies we identified the Teflon membrane as limiting factor for a fast response time [4], thus we executed further experiments using different filter configurations in order to eventually design and test a novel filter package comprising Teflon filters of different pore size. In Fig. 2, the various filter configurations tested in this study are sketched. The novel filter package consists of a zeolite filter (19), and two Teflon filter disks (16b) with pore size of 10 µm (diameter: 6 mm and thickness: 1 mm). The filter package is located between the sensor element, i.e., quartz oscillator (top) and the gas input (bottom).

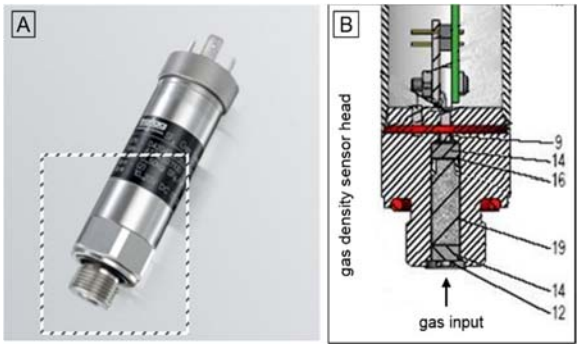

Fig. 1. Gas density sensor based on a quartz tuning fork oscillator (8774, Trafag, Switzerland): (A) Photo of sensor head, (B) Cross section of the front part (dashed section in (A)). The standard filter package between quartz oscillator location (9) and gas input consists of a metal clamp (12), two metal sinter filters (14), a zeolite filter (19), and a Teflon filter membrane (16).

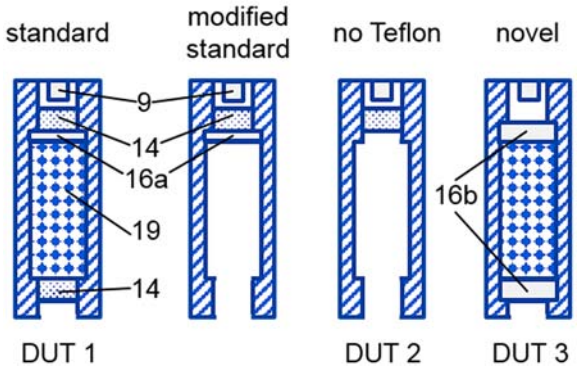

Fig. 2. Sketch of tested filter packages. Sensor element (9), metal sinter filter (14) and Teflon filter (1 µm pore size (16a), 10 µm pore size (16b)), zeolite filter (19). Open to gas at the bottom.

In the following we describe the materials and procedures used and show our experimental setup (Section 3) using sensors of different filter configuration both for gas mixtures (nitrogen plus perfluoroketone) and for pure gas (nitrogen, sulfur hexafluoride). The dependence of the gas diffusion (and thus of the response time) on the filter package (and in particular on the Teflon filter pore size) is reported in Section 4.

3. Materials, Setup and Procedure

The devices under test are Trafag density sensors (type 8874 (analogue interface) and 8875 (digital interface)) with different filter package configurations. The filter configurations comprise the “standard”, the “modified standard”, the “no Teflon” and the “novel” filter as shown in Fig. 2.

The test gas used for the first series of experiments is a gas mixture composed of light and heavy gas molecules, i.e., nitrogen (M = 28 g/mol) and perfluoroketone (PFK) (M = 266 g/mol) [5]. The experimental setup with test vessel and attached sensors is shown in Fig. 3 and is described in detail elsewhere [6]. The experiments were performed having the gas vessel and sensors in a temperature controlled chamber at a temperature of T = 23.5 °C. To monitor the pressure in the gas vessel during evacuation and filling a *p*-*T*-sensor (Keller PAA-33X) was used as a reference. Before gas filling, the gas vessel was evacuated down to a pressure of a few millibars. The gas was already a homogeneous mixture before filling. The signals were read out via Modbus RS485 interface and displayed, averaged and recorded using a dedicated LabView software.

Fig. 3. Cylindrical gas vessel with Trafag density sensors (orange cable connector) and pressure sensor attached in front of a temperature control chamber.

The test gas used for the second series of experiments are pure gases, on one hand nitrogen on the other hand sulfur hexafluoride (M = 146 g/mol). The experimental setup with the gas manifold, control valves, three attached density sensors, one reference pressure sensor and vacuum pump is shown in Fig. 4. The temperature conditions during the test were 22 °C < T < 24 °C.

The output signals are recorded over precision shunt resistors of 100 Ω. Thus, a current output of 4 mA – 20 mA corresponds to 0.4 V – 2.0 V. The analog voltage signals are logged on a PC.

The response time definitions we used are shown in the simplified sketch in Fig. 5. The graph shows two sensor responses upon gas exposure (at $t = t_0$), which we typically observe in our experiments: One curve is monotonically increasing (blue) and the other one is not monotonic (red). For the results in Section 4.1, the criterion for stationary signal condition, i.e., $S(t > (t_0 + t_{100})) \approx$ const., was a slope close to zero, i.e., $|dS/dt| < 0.01$ Hz/h and $|dS/dt| < 0.005$ Hz/min., for sensors with large response times and for sensors with small response times, respectively. For the case of non-monotonic sensor response (red curve in Fig. 5), those intersections which are closer to t_{100} are considered for the determination of t_{90} and t_{99}. Using the red curve as example: Once t_{100} is determined using the criterion described above, the larger time point (t_2) is taken to determine t_{90}, i.e., $t_{90} = t_2 - t_0$, whereas $S(t_1) = S(t_2) = 110$ and $t_2 > t_1$.

Fig. 4. Manifold with 3 Trafag density sensors (black cable connectors), arranged in a T-configuration, control valves, pressure sensor and vacuum pump (right).

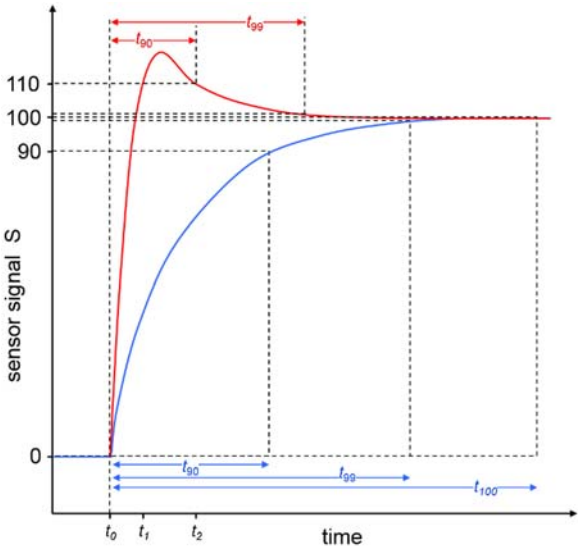

Fig. 5. Sketch of sensor signal response illustrating response time definitions. At stationary conditions the sensor signal is normalized to 100.

4. Results

4.1. Gas Mixture

In order to discriminate effects of the filter package on diffusion times of large molecules (perfluoroketone) from effects on small molecules (nitrogen) we first tested the response time of the density sensor on exposure to pure nitrogen. The response of a sensor with standard filter package to N_2 is shown in Fig. 6A. The graph shows the time coarse

of temperature normalized pressure and density during a pressure change from $p \approx 0.005$ bar to $p \approx 5.9$ bar. The shown frequency signal f of the density sensor can be converted to density as described in [3].

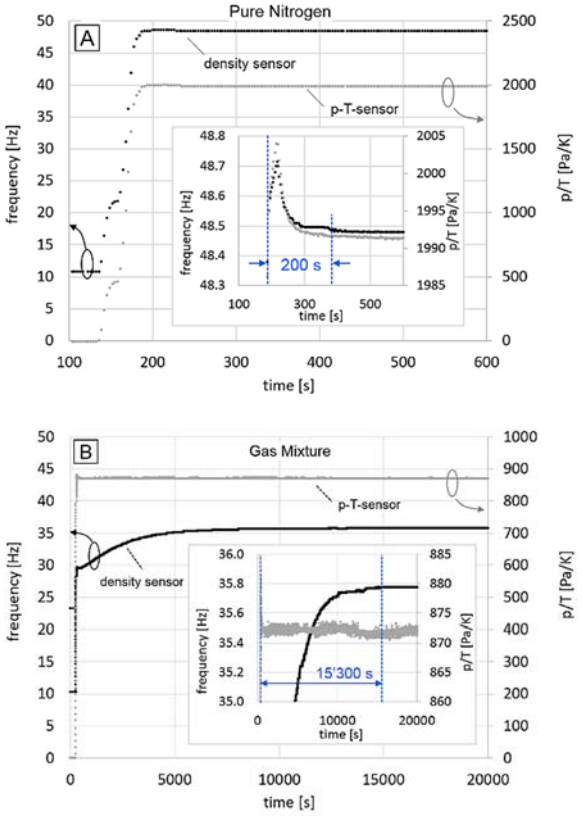

Fig. 6. Response of a sensor with standard filter package to a sudden change in gas pressure. (A) Pure nitrogen, (B) Mixture of nitrogen (95 %) and perfluoroketone (5 %).

Note that we aim at a precision of $\Delta\rho \approx 5$ g/m^3 corresponding to $\Delta f \approx 0.05$ Hz. The inset of Fig. 6A shows a f-zoom of the same graph. We obtain for pure nitrogen a response time of $t_{100} = 200$ s, not much different from the p/T signal that stems from the auxiliary p-T-sensor. Thus, the filter package of the density sensor does not cause significant response delay in the case of N_2. The small pressure and density swing (13 Pa/K and 0.2 Hz) in the time between 150 s < t < 300 s is most likely due to a short temperature non-equilibrium in the sensors due to adiabatic heating of the gas during filling.

The behavior of the response time is fundamentally changed upon exposure to the gas mixture of nitrogen and perfluoroketone (composition 95:5), shown in Fig. 6B. In a first response the density sensor shows a fast partial response (frequency change from $f = 10$ Hz to $f = 30$ Hz). This fast response can mainly be attributed to the fast diffusion of N_2 through the filter package. The following slow diffusion of the large perfluoroketone molecules lasts for 4.25 h until steady state conditions are reached. We obtain for the mixture a response time of t_{100} = 15'300 s (inset of Fig. 6B). In contrast to the diffusion limited density sensor signal, the p/T signal from the p-T-reference sensor which is proportional to the density (for ideal gas law approximation) is constant already 50 s after gas filling.

In Fig. 7 we show the response of two density sensors with different filter configuration exposed in the same experiment to the N_2/PFK (95:5) gas mixture (the "standard" filter and the "no Teflon"). The graph shows the transition from $p \approx 0.005$ bar to $p \approx 4.5$ bar. Fig. 7B and Fig. 7C display a f-zoom of the graph in Fig. 7A. We observe a t_{100} response time of 33'000 s and 300 s for the "standard" filter and for the "no Teflon" filter, respectively. It is evident that by omitting the Teflon filter the t_{100} response time of the density sensor can be improved by two orders of magnitude.

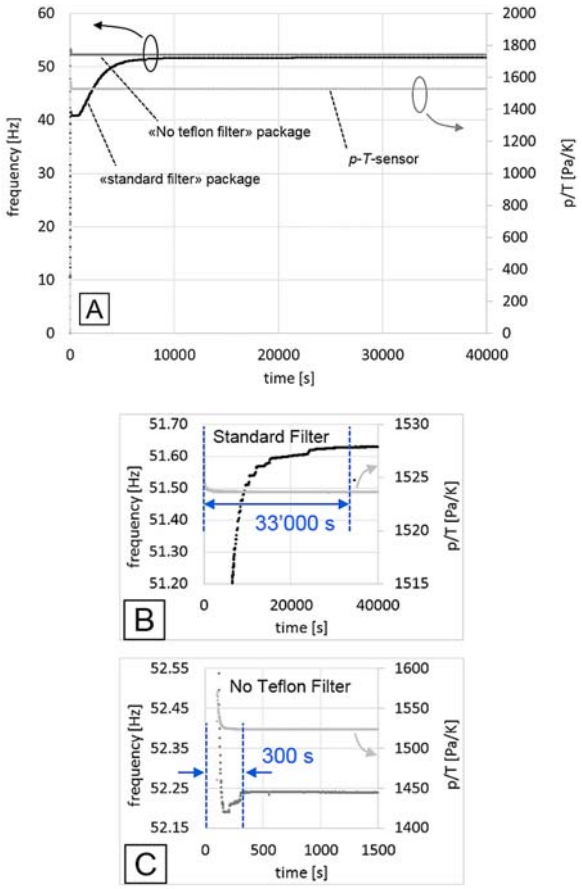

Fig. 7. Response time of two density sensors of filter package "standard" and "no Teflon" to a sudden change in gas pressure. (A) Overview plot, (B) f-zoom of plot (A) and sensor with "standard" filter package, (C) f-zoom of plot (A) and sensor with filter package "no Teflon".

In order to exclude significant effects on response time from the zeolite filter beads, we exposed the filter package "modified standard" to the gas mixture (Fig. 8). Also here, we observe a large t_{100} response time of t_{100} = 24'000 s.

We can conclude that the response time of the density sensor with filter packages where the Teflon filter membrane is integrated ("standard" and

"modified standard") is diffusion limited and is approximately two orders of magnitude larger than in the "no Teflon" filter configuration (Table 1). This can be explained by the large molecular size and heavy mass of the perfluoroketone, that has a much higher diffusion time through the Teflon filter compared to the nitrogen molecule. We can conclude from the observations that the other components of the filter package, such as zeolite filter beads and metallic sinter filter have a minor influence on the t_{100} response time. The question how large the influence of the Teflon pore size in a Teflon filter affects the response time is addressed in Section 4.2.

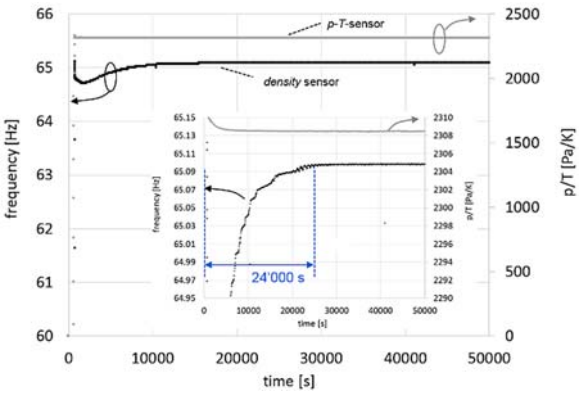

Fig. 8. Response of a sensor with "modified standard" filter package to a sudden change in gas pressure. Mixture of nitrogen (96.5 %) and perfluoroketone (3.5 %).

Table 1. Response time t_{100} [s] of density sensors with different filter configurations ("standard", "modified standard", "no Teflon") exposed to pure N_2 and a mixture of PFK and N_2. ((1), (2) Results of two similar experiments: Fig. 6B(2), Fig. 7B(2)).

	[s]	Standard	Modified standard	No Teflon
N_2	t_{100}	200	-	-
SF_6	t_{100}	15'300(1) 33'000(2)	24'000	300

4.2. Pure Gas

The knowledge of response delay for large gas molecules due to the Teflon filter membrane led to further investigations with a novel filter package design using a Teflon filter with significantly larger pore size ("novel" or DUT 3 in Fig. 2). We tested this novel filter package in comparison with two other filter packages ("standard" or DUT 1 and "no Teflon" or DUT 3) by analyzing the t_{90} and t_{99} response times upon exposure to SF_6 in various different experimental preconditions.

Firstly, Fig. 9 shows the signal response of the three different devices under test (DUT) upon exposure to pure N_2. The manifold was evacuated for 4 minutes. Afterwards, a N_2 pressure of 8.8 bar was applied. The response t_{90} is almost immediate within a few seconds. The response time t_{100} is in the order of several hundred seconds which is consistent with our results in Fig. 6A. All DUTs show some signal overshoot reaction in the percent range (Fig. 9B), among them DUT 2 with the strongest overshoot. After less than 30 s, DUT 1 and DUT 3 are within 1 % of the final value. In conclusion, for N_2 we observe a fast response for all filter configurations.

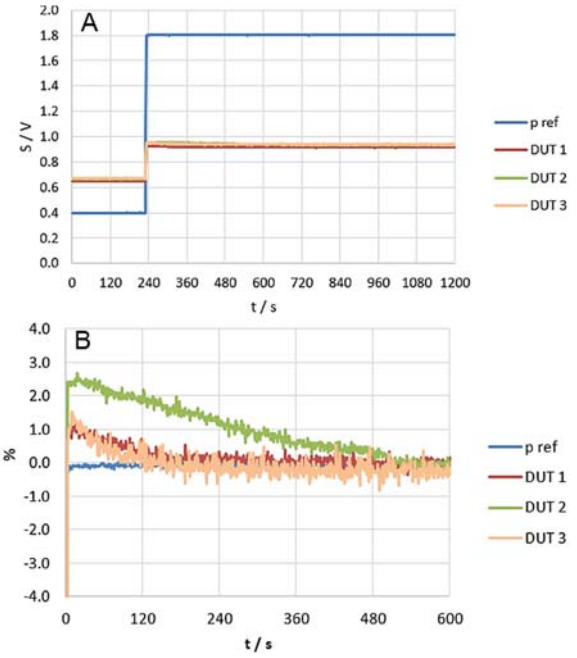

Fig. 9. Response of density sensor with 3 different filter packages to a sudden change in gas pressure (N_2). (A) Overview, (B) Zoom of (A): Relative deviation of signal from signal value at equilibrium ($t > 1000$ s), plotted from $t = 230$ s onwards.

Fig. 10 shows the signal response of the three different DUTs upon exposure to pure SF_6. After the manifold with the sensors attached was evacuated for one minute, an SF_6 pressure of 6.3 bar was applied. It is evident that the response time of the sensor with "novel" filter (DUT 3) is significantly faster than for the sensor with "standard" filter (DUT 1). The fastest response is shown by the density sensor without filter ("no Teflon" or DUT2). See Table 2 for comparison of response times. In conclusion, when the density sensors are exposed to SF_6 the response time t_{90} can be improved by a factor of two using the "novel" filter package and t_{99} can likely be improved by a factor of ten.

In real applications, when the sensor will be attached to an SF_6 filled compartment, the small cavity, where the filters and the quartz sensor are located, is typically exposed to environmental air prior to attachment. After attachment of the sensor to a gas vessel under test, the air at the sensor location has to be displaced completely by the analyte gas, e.g. SF_6, in order to obtain an accurate density reading.

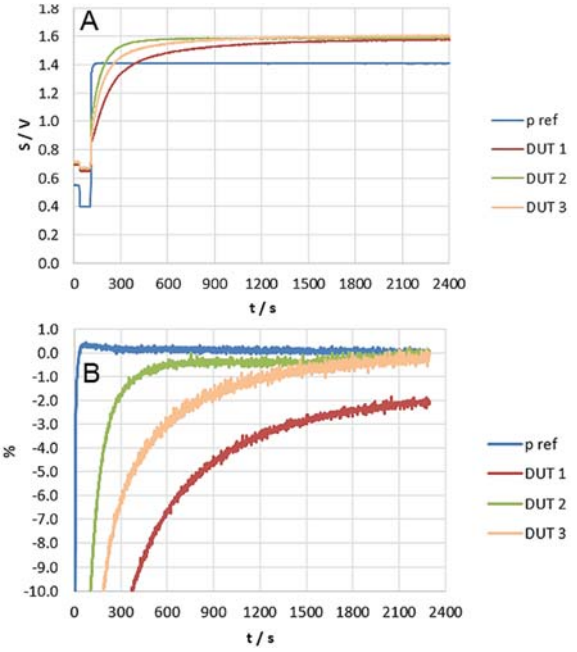

Fig. 10. Response of density sensor with 3 different filter packages to a sudden change in gas pressure (SF_6). (A) Overview, (B) Zoom of (A): Relative deviation of signal from final signal value ($t > 2400$ s), plotted from $t = 110$ s onwards.

Table 2. Response time t_{90} and t_{99} [s] of density sensors with different filter configurations exposed to pure N_2 and to pure SF_6.

	[s]	Standard/ DUT 1	No Teflon/ DUT 2	Novel/ DUT 3
N_2	t_{90}	1	1	1
	t_{99}	20	280	30
SF_6	t_{90}	375	105	190
	t_{99}	> 6000	430	1200

We quantitatively investigated this diffusion limited process using the three different filter packages in two experiments, one with 1 bar environmental air in the filter package and the other one with SF_6 in the filter package. The results are shown in Fig. 11 and Fig. 12. In the experiment of Fig. 11 the three density sensors were not evacuated prior to attachment to the manifold, but rather exposed to air and then attached to the SF_6 filled manifold. In general, the sensors show a much larger response, both for t_{90} and for t_{99} (see Table 3), compared to the experiment of Fig. 10.

In the experiment of Fig. 12 the three density sensors were not evacuated prior to attachment to the manifold, but rather filled with 6 bar SF_6 and left exposed for the time duration of 2 h. Afterwards they were disconnected from the SF_6 and exposed for one minute to ambient air and then connected to the manifold which was filled with SF_6. The graphs of Fig. 12 show that the time characteristics is very different from Fig. 11. The sensors show a much faster response, both for t_{90} and for t_{99} (see Table 3). Besides the novel filter package design for density sensors for improving the time response, also the storage of these sensors could be revised, i.e., for applications where time response after sensor attachment is critical, the sensors could be stored in the atmosphere of the analyte gas.

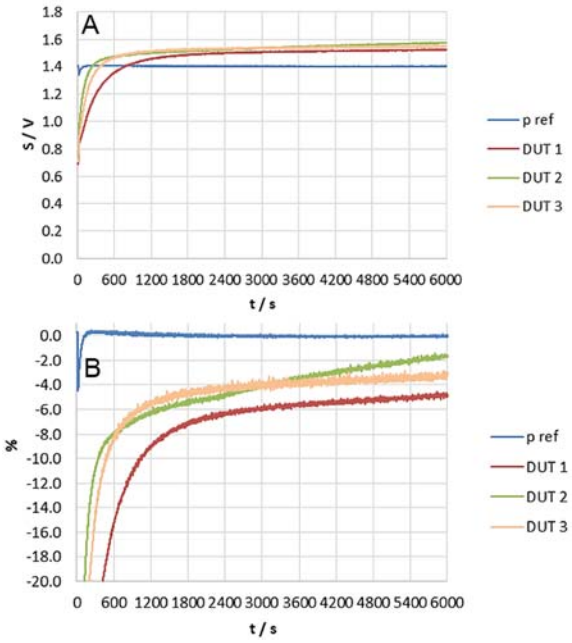

Fig. 11. Response of density sensor with 3 different filter packages to a sudden change in gas pressure (SF_6). The sensor input was filled with 1 bar environmental air prior to connection to the gas manifold. (A) Overview, (B) Zoom of (A): Relative deviation of signal from final signal value at equilibrium.

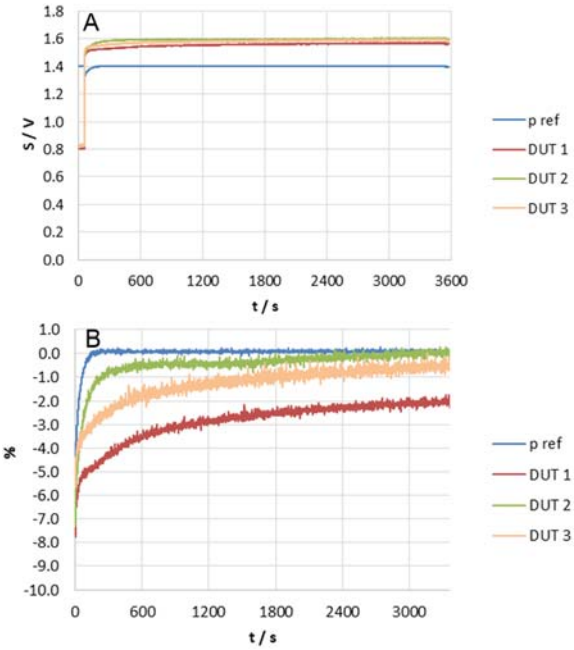

Fig. 12. Response of density sensor with 3 different filter packages to a sudden change in gas pressure (SF_6). The sensor input was preconditioned with SF_6 prior to connection to the gas manifold. (A) Overview, (B) Zoom of (A): Relative deviation of signal from final signal value at equilibrium.

Table 3. Response time t_{90} and t_{99} [s] of density sensors with different filter configurations exposed to pure SF_6 with different gas prefilling of the sensor head prior to exposure to the analyte gas.

	Sensors prefilled with	[s]	Standard/ DUT 1	No Teflon/ DUT 2	Novel/ DUT 3
SF_6	air	t_{90} t_{99}	1060 > 6000	446 > 6000	490 >6000
SF_6	SF_6	t_{90} t_{99}	2 > 6000	1 250	1 1750

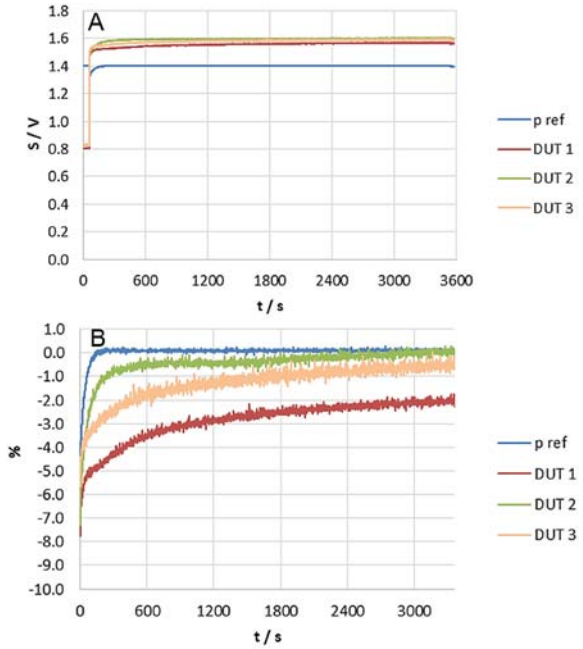

Fig. 12. Response of density sensor with 3 different filter packages to a sudden change in gas pressure (SF_6). The sensor input was preconditioned with SF_6 prior to connection to the gas manifold. (A) Overview, (B) Zoom of (A): Relative deviation of signal from final signal value at equilibrium.

7. Conclusions

In conclusion, we investigated and quantified the response time of commercial gas density sensors with the goal to introduce a novel sensor package that significantly reduces the response time. Our experiments showed that a specific element, the Teflon filter membrane (pore size 1 μm), in the standard filter protection package, limits significantly the diffusion rate of large gas molecules. These findings led to a revision of the sensor package resulting in the replacement of the Teflon membrane by two Teflon filters of larger pore size (10 μm). We demonstrated that sensors of the novel filter design, applied to electrical insulation gas, show a response time improvement of a factor of two for the t_{99} response time and one to two orders of magnitude for the t_{100} response time. Thus, a novel superior filter configuration was designed which will find its way into our future density sensor products. This novel sensor technology is particularly attractive to be used for improved emission monitoring and servicing of gas- and gas-mixture-insulated electrical switchgear and possibly will leverage the introduction of environmentally friendly gas mixtures which represent alternatives to SF_6.

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