Isoprene Selective Gas Sensing by Adsorption Filter

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Abstract:
Isoprene is a promising breath marker for the non-invasive monitoring of high blood cholesterol. However, portable gas sensors (e.g. metal-oxide based), do not offer the required selectivity to detect isoprene levels in complex human breath. Here, we present a new filter concept enabling fast and highly selective detection of isoprene at the ppb level and high humidity. The filter is a packed bed of activated alumina powder placed upstream of a non-specific Pt-doped SnO$_2$ sensor. Hydrophilic breath compounds (e.g. acetone, ammonia, ethanol, methanol) are adsorbed and retained on the adsorbent during detection of hydrophobic isoprene, which is not affected by the filter. This way, isoprene is measured within seconds in simulated breath with unprecedented selectivity >100 to the tested analytes. Full regeneration of the filter within 10 min by purging with air enables continuous application, promising for portable breath isoprene analyzers.

Key words: filter, activated alumina, breath analysis, cholesterol, SnO$_2$, flame spray pyrolysis

Introduction
Exhaled isoprene is a by-product of the cholesterol biosynthesis [1] with typical breath concentrations ranging from 22 to 234 ppb. The detection of isoprene in breath is thus very promising for the non-invasive measurement and monitoring of high blood cholesterol, the major cause of cardiovascular diseases leading to millions of deaths every year.

Nanostructured metal-oxide (MOx) gas sensors are attractive for breath analysis by being inexpensive, simple-in-use, portable and they have been applied already for non-invasive fat burn monitoring during exercise and rest [2]. MOx sensors, however, typically lack selectivity to detect target analytes in breath, especially when these occur at low concentration. This can be solved in some cases by material design (e.g. Si:MoO$_3$ for NH$_3$ [3]) or membranes (e.g. for formaldehyde [4]). For isoprene, highest selectivities achieved so far are in the range of 4-15 for acetone, ethanol and ammonia by Ti:ZnO [5]. However, this is not enough for precise isoprene detection in breath.

Here, this is solved by placing a filter upstream of a non-specific gas sensor that exploits the hydrophobicity of isoprene in contrast to other major breath components. The filter is a packed bed of activated alumina featuring high porosity and surface area (155 m$^2$/g). It adsorbs and retains hydrophilic compounds while isoprene passes unhindered and is detected by the sensor with outstanding selectivity (inset Fig. 1b).

Experimental
1 g activated alumina (50-300 mesh, 155 m$^2$/g) inside a Teflon tube (Ø 15 mm) is placed upstream of a non-specific Pt-doped SnO$_2$ sensor prepared by flame spray pyrolysis [4] and heated to 400 °C by a Pt back heater. Analytes are admixed with synthetic air at 90% RH in a mixing setup adapted from [3] and supplied through the filter to the sensor at 1 L/min.

Results
The Pt-doped SnO$_2$ sensor shows fast response time and high sensitivity to 500 ppb of isoprene at 90% RH. However, it is non-specific, thus cannot distinguish between isoprene and other breath-relevant analytes. This becomes evident when plotting the sensor response (Fig. 1a) when exposed to 500 ppb of isoprene (blue), acetone (red), ethanol (orange), methanol (green) and ammonia (purple).

Consequently, this sensor cannot measure isoprene selectively in breath mixtures.

When adding the activated alumina filter (Fig. 1b) an identical response for isoprene is obtained with barely affected response dynamics, while all hydrophilic analytes are held back. This results in unprecedented high isoprene-selectivity (>100) to all analytes for up to 40 s, clearly outperforming state-of-the-art chemoresistive isoprene sensors. Only after, the sensor starts to detect acetone while it takes even longer for the other hydrophilic analytes.
The breath is composed of >800 compounds, so evaluation of the filter–sensor system in gas mixtures is crucial. For this, it was exposed to simulated breath pulses for 30 s containing breath-relevant concentrations of isoprene (106 ppb), acetone (477 ppb), methanol (461 ppb) and ammonia (833 ppb) to simulate a breath-isoprene measurement. The results are shown in Fig. 2 where the sensor response is plotted for three such consecutive pulses, resulting each in two distinct responses. The first one increases immediately at the start of the 30 s exposure and is stable during it. It is attributed to isoprene, giving the same response in single gas measurements. All hydrophilic analytes are held back by the filter and only later result in a second response, which is delayed and spread. This enables precise measurement of isoprene despite the more concentrated interferants. By purging with air, the filter fully regenerates within 10 min and can be used for the next measurement.

As a result, a simple packed bed filter of 1 g commercial activated alumina turned a non-specific gas sensor isoprene selective. The filter does not change the sensor response to isoprene while the influence of hydrophilic interferants is eliminated. Combined with a nanostructured Pt-doped SnO\textsubscript{2} sensor, this leads to the detection of isoprene down to 5 ppb within seconds ($t_{90} = 2.9$ s) with selectivity >100 to major other breath markers. The filter–sensor system showed stable performance with reproducible (regeneration within 10 min) and accurate isoprene detection in simulated breath mixtures. This filter is modular and, based on its small size and low price, can be readily integrated into inexpensive and portable breath analyzers promising for non-invasive monitoring of blood cholesterol levels.

References


