SKIN-EMITTED ACETONE DETECTION TOWARD SELF-MONITORING OF FAT METABOLISMS
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ABSTRACT
We aimed to develop a device for detecting very low concentrations of skin-emitted acetone (skin acetone) toward self-monitoring of fat metabolisms in our daily life. Skin acetone analysis is expected to be a powerful tool to prevent and alleviate lifestyle-related diseases, however existing methods lack in repeated use and require large-sized apparatus. We propose to use a zeolite array as a concentrator of skin acetone to relieve the requirements for the detector sensitivity, and showed that it can produce more than tenfold concentrated skin acetones. Our results will help to open the way for a wearable skin acetone analyzer.

KEYWORDS: Skin gas, Health monitoring, Wearable device, Zeolite array, Acetone concentrator

INTRODUCTION
Acetone is a metabolic product of the breakdown of body fat, produced in the blood, that is exhaled through our breath and skin with very low concentrations [1, 2]. It is known that breath and skin acetones are good indicators of fat metabolisms which associate with diet, aerobic exercise, obesity, and diabetic control [2]. Skin acetone analysis is advantageous compared to breath acetone analysis thanks to its nature of unconscious and continuous emission from our skin surface. This means that skin acetone analysis requires no active action such as deep exhalation and could be achieved more precise quantification that excludes intentionally factors such as the flow rate of exhalation. If we can devise a wearable skin acetone analyzer, it could be a powerful tool to prevent and alleviate lifestyle-related diseases. However, existing methods that employ skin gas collection bags [2] and trapping filters [3] lack in repeated use, and require large-sized apparatus such as gas chromatography to detect skin acetones with high sensitivity.

Here, we propose to use zeolite as a concentrator of skin acetone to relieve the requirements for the detector sensitivity (Figure 1). Zeolite is a porous material and is expected to adsorb and desorb acetone molecules selectively by designing of its pore size and hydrophobicity. If we can detect the concentrated skin acetones by using semiconductor-based gas sensors, it will help to open the way for a wearable skin acetone analyzer. The aims of this work were to investigate the basic performance of our zeolite device and to confirm the feasibility of the device for the pure acetone gas as well as real skin-emitted acetone gas.

EXPERIMENTAL
We first tried to fabricate a zeolite array that will be suitable for the immediate heating and cooling on a silicon substrate. An acrylic mold with periodic 500 μm-square pillars was fabricated by stereo lithography (Perfactory 3 SXGA+ Mini, EnvisionTEC GmbH, Germany) and polydimethylsiloxane (PDMS) solution was deposited on the mold and baked (Figures 2(a) and 2(b)). Then the fabricated PDMS mask with periodic 500 μm-square holes was peeled away from the mold (Figure 2(c)) and was treated with oxygen plasma (Figure 2(d)). The hydrophilic PDMS mask was stuck to a silicon substrate (Figure 2(e)) and zeolite slurry was deposited on the mask (Figure 2(f)). We chose HSZ-390HUA zeolite (Tosoh Corp., Japan) as a concentrator of skin acetone because its pore size is comparable to the diameter of acetone molecules and its hydrophobicity is high. The deposited zeolite slurry was penetrated through the PDMS mask by deaeration in a vacuum desiccator (Figure 2(g)) and the mask was peeled away from the silicon substrate (Figure 2(h)). The obtained array was calcined at 800°C for one hour by using an electric furnace (Figure 2(i)).
To investigate the basic functionalities of the zeolite array, we generated pure acetone gas in a closed space with 16.9 mL of a glass vial container and determined how much amount of acetones is adsorbed/desorbed into/from the zeolite array by using a conventional gas chromatography (SGEA-P2, FIS Inc., Japan). Similarly, we examined if the zeolite array can adsorb/desorb acetones repeatedly without degrading its performance. To further investigate the feasibility of the zeolite array, we analyzed a magnitude of concentration effect for skin acetones emitted from a human left palm at different skin gas collection time.

RESULTS AND DISCUSSION

We fabricated a zeolite array on a 7 mm-square silicon substrate as described above (Figure 3(a)) and microscopic observation confirmed that 500 μm-square and spacing zeolite array was successfully obtained (Figure 3(b)). We next examined adsorption and desorption characteristics of the obtained zeolite array for pure acetone gas. We found that 81% of generated pure acetones was adsorbed into the zeolite array and 94% of the adsorbed acetones was desorbed from the zeolite array in response to the thermal stimulus (Figure 4). This adsorption-desorption processes were able to repeat at least 12 times; confirming that the zeolite array can adsorb and desorb acetones repeatedly without degrading its performance (Figure 5).

Figure 3: (a) Photograph of the zeolite array on a 7mm-square silicon substrate. (b) Microscope image of the zeolite array. The scale bar corresponds to 500 μm.

Figure 4: Adsorption and desorption characteristics of the zeolite array for pure acetone gas. The error bars represent the standard deviation from three distinct measurements.
We finally investigated adsorption and desorption characteristics of the obtained zeolite array for real skin-emitted acetone. Skin acetones were collected from a human left palm by creating a closed space containing the zeolite array inside of a glass vial container (Figure 6, Inset) and the amount of skin acetones desorbed from the zeolite array was analyzed under the different skin gas collection time. The amount of naturally emitted skin acetones per minute was averaged from the 30 minutes observation. The result showed that the zeolite array was able to adsorb and desorb skin acetones and was able to produce more than tenfold concentrated skin acetones compared to the natural emission (Figure 6). Our proposed method is suitable for the wearable analyzer because adsorbed skin acetones are retained in the zeolite array even if the closed space for skin gas collection is temporary collapsed due to the activities (accompanied with twist and elastic movement of the skin surface) in our daily life.

**CONCLUSION**

We succeeded in fabricating a zeolite array on a silicon substrate, and found that the zeolite array can adsorb skin acetones and can desorb them in response to thermal stimulus; resulted in producing more than tenfold concentrated skin acetones. Since this magnitude will be enough to detect by semiconductor-based gas sensors and the adsorption-desorption processes were able to repeat without degrading its performance, our results will help to open the way for a wearable skin acetone analyzer toward self-monitoring of fat metabolisms in our daily life.

**REFERENCES**


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