Multiplexed Graphene Sensors for Detection of Ions in Electrolyte

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Sponsorship: MIT-ARL ISN and NSF CIQM

In the era of Internet-of-things (IoT), new types of sensors are developed everyday to gather relevant data in daily activities. Wearable electronics such as sweat sensors targeting informative biomarkers have been heavily investigated. However, these electronics mostly only contain one sensor for each type of analyte and the performance is evaluated and optimized separately. When applied to real-world application with complex environment, the reproducibility and the reliability of such device is questionable. Here we present a platform technology for multiplexed, large-area sensing array for more reliable measurement. Graphene is used as signal transducer because of its high surface-to-volume ratio and excellent electrical properties. By utilizing a material jetting 3D printer, we are able to deposit different types of functionalization on specific regions of the array to achieve multiplexed sensing. Here we demonstrate a fully integrated sensing array with three types of ion-selective membranes (ISMs) to achieve detection of sodium, potassium and calcium as shown in Figure 1. We chose these ions because their physiological importance. Each types of functionalization covers over 70 working devices and in total more than 200 devices are functional in one array.

The multiplexed sensor array is first tested with various concentration of solutions contain pure K, Na or Ca ions. All three types of sensors show excellent Nernstian sensitivity, high reversibility and fast response time towards their target ion. They also exhibit a moderate level of sensitivity towards other two types of ions. By having an array of non-perfectly selective sensors, more information is available for analysis. Using Principle Component Analysis on the sensor array output, we are able to cluster and then identify the type of ion in the tested solutions as shown in Figure 2. The sensor array is also tested with a set of mixture solutions that are prepared by fixing the concentration of interfering ions while varying concentration of a specific type of ions. Similar clusters are observed indicating the sensor array's ability for identifying which type of ion concentration is changing within a complex mixture solution. This work demonstrates the possibility of achieving highly reliable multiplexed sensing array that can be deployed in complex environments. By collecting data from a statistically significant sample size, we would be able to apply more sophesticated statistical methods or machine learning models to further associete complex mixtures for real-world applications.

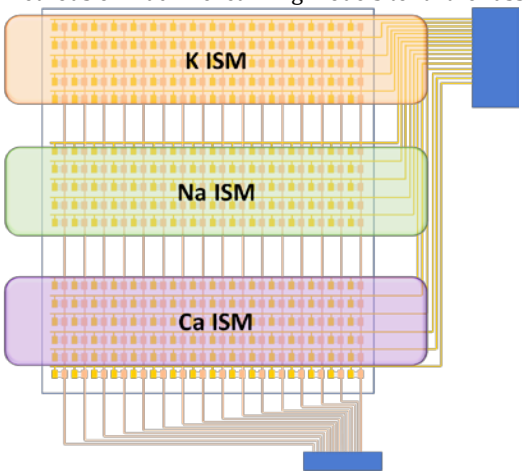

Figure 1: Schematics for the sensor array. Three types of ion-selective membranes are deposited to achieve multiplex sensing

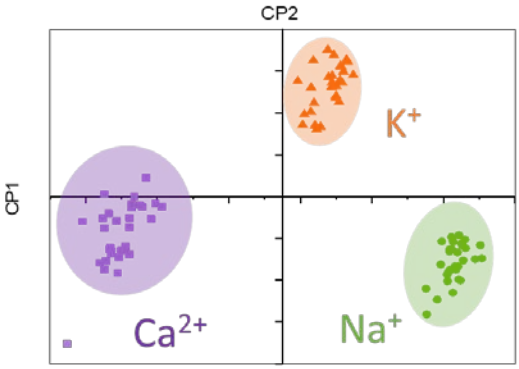

Figure 2: Principle component analysis of the sensor array with pure solution of potassium, sodium or calcium ions

Further Reading

[1] C. Mackin et al., "Chemical sensor systems based on 2D and thin film materials," 2D Mater., Jan. 2020.
[2] W. Gao et al., "Fully integrated wearable sensor arrays for multiplexed in situ perspiration analysis," Nature, vol. 529, no. 7587, pp. 509–514, 2016.
[3] J. Kim, A. S. Campbell, B. E. F. de Ávila, and J. Wang, "Wearable biosensors for healthcare monitoring," Nature Biotechnology, vol. 37, no. 4. Nature Publishing Group, pp. 389–406, 25-Apr-2019.