

# Graphene Based Flow Sensors

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We report on attempts to produce a graphene based liquid flow sensor. Our results indicate that modifications of the electric double layer, formed in the vicinity of the graphene surface, dominate over mechanisms responsible for liquid flow-induced voltage/current generation. Several graphene structures were tested in different measurement configurations, aimed to maximize the generated signal amplitude and its stability. Some realizations of working devices in water as well as in aqueous solutions of NaCl or HCl are presented.

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## 1. Introduction

Graphene is a chemically and mechanical resistant material and can therefore readily be used for applications in harsh environments. This makes graphene, also due to its excellent electrical properties, a very promising material for the use in medicine, chemistry, food industry as well as in electronics, optoelectronics, robotics and nanotechnology. One of the areas of research, which is currently being widely investigated, is the use of graphene as a liquid flow sensor [1–4]. The reports addressing such sensors, which were published so far, are very contradictory [5, 6]. This indicates that the underlying physics is still not well understood and that more in-depth studies are needed in order to shed more light on this subject

## 2. Theory of double layer

A double layer consists of two layers of oppositely charged ions (see Fig. 1) formed at the interface of a material immersed in liquid [7]. The first layer close to the surface consists of ions (positive or negative) adsorbed due to chemical interactions. In this layer, the ions are densely packed and tightly attached to the surface. The second layer is created via the Coulomb interaction with the first layer and consists of oppositely charged ions, which screen the first layer potential. This layer is much more loosely bound to the surface, and can be influenced by ions moving in the liquid.

## 3. Result and discussions

Different approaches were used to observe generation of flow induced current/voltage in graphene samples. Many of them were found not to work effectively. Some of them like the use of Cu as gate (instead of Pt) or the use of Cr as metallization, provided some results in voltage/current generations but their origins seem to be mostly due to electrochemical processes taking place at the interface between graphene and/or metallization and the solution. We checked that graphene flakes transferred to

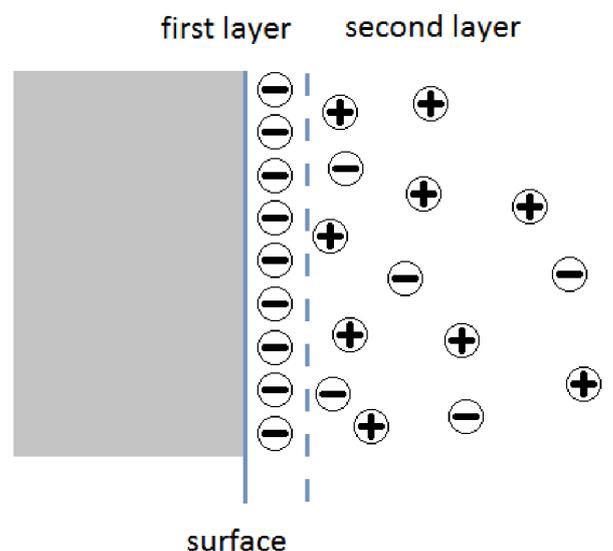


Fig. 1. Distribution of charges in the double layer.

SiO<sub>2</sub> and PET gave worse results than epitaxial graphene grown on SiC. We found that a passivation of metallic contacts performed using SiO<sub>2</sub> did not significantly alter the current/voltage generation efficiency. The SiO<sub>2</sub> passivation layer showed, moreover, rapid degradation. Besides many failures we found some ways that significantly improved the quality of the results. The simplest system used in our experiments contains only one electrode which served as the polarizing gate.

Much better results, with respect to the signal to noise ratio as well as signal stability, were obtained by using a Pt gate and a Ag/AgCl reference electrode simultaneously (Fig. 2). Depending on the type of graphene (*p*-type, *n*-type, monolayer, multilayer) and the type of liquid (deionized water, aqueous solution of NaCl, HCl, or ethanol) different experimental conditions were found to be optimal. In some cases better sensitivity to the liquid flow was obtained by applying a voltage between

the sample and the gate. One of the first things we noticed was the need to separate the contact pads from each other. At the beginning we performed this electrical isolation by etching a part of the graphene. Later we achieved this by performing a mechanical scratch mark between the pads (Fig. 2). In order to exclude possible effects on the generated signals due to the metallization, we constructed a sample for which the metallic contacts, passivated with an epoxy resin, were not immersed in the liquid. In this case a part of the graphene layer was separated by a scratch, thus serving as electrical leads and as a active part of the flow sensor (Fig. 2).

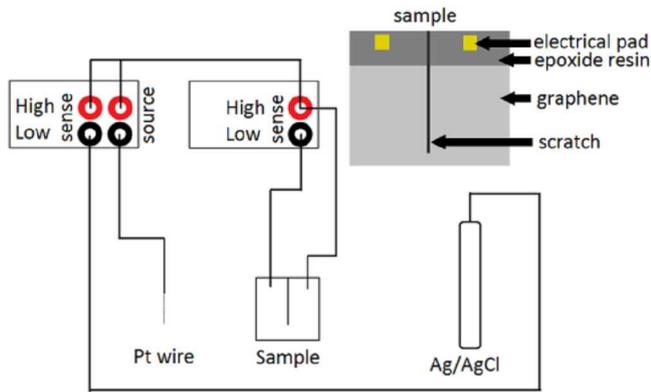


Fig. 2. Electrical wiring diagram of the most common operating mode with the use of a Pt wire and an Ag/AgCl electrode and scheme of the sample.

In Fig. 3 we present an example of the obtained results. In order to check the influence of the amount of the immersed surface, experiments with different immersion levels were performed. As can be seen in Fig. 3, both the offset and the amplitude of the generated current increase with deeper immersion level.

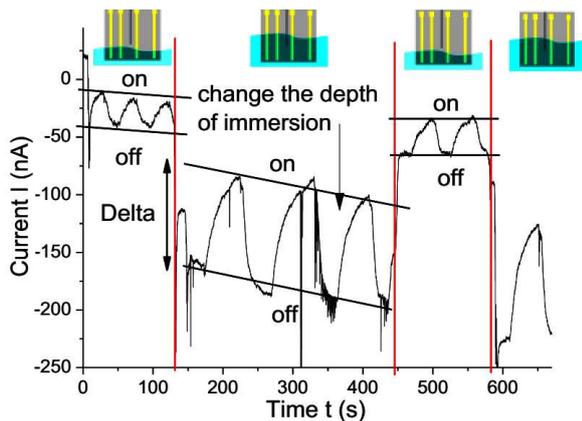


Fig. 3. The difference in response for electrical connections immersed and not immersed in the liquid. Changes in amplitude are caused by turning on and off the flow. Lines with the caption "on" indicate the established signal level when there is a fluid flow; lines with the caption "off" show the established signal level in the absence of a fluid flow.

Surprisingly, we found that the observed signals are dominated by a component which is not dependent on the direction of fluid flow.

We have performed measurements for different aqueous solutions. Figure 4 shows how the generated signal depends on the aqueous HCl concentration. It was observed that the generated signal increases linearly upon concentration increase for concentrations larger than about 4%. In the case of other aqueous solutions the situation is more complicated. In particular for large concentrations of organic liquids the signal tends to zero. However, by applying an optimal gate voltage we obtained efficient flow sensitive signals up to a concentration of about 40% of an aqueous solution of ethanol. Despite many prob-

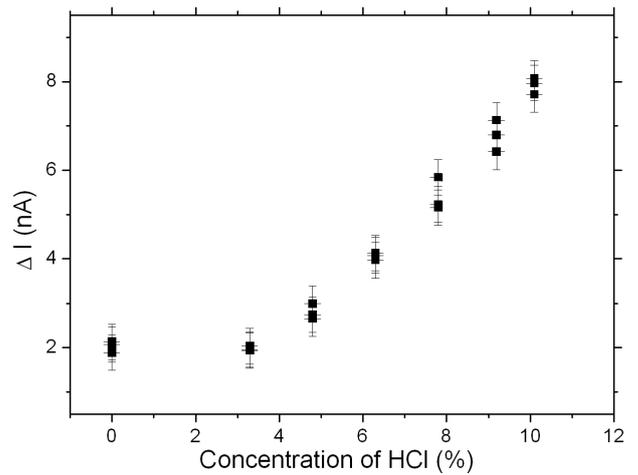


Fig. 4. Flow induced current measured for different HCl concentrations in the water.

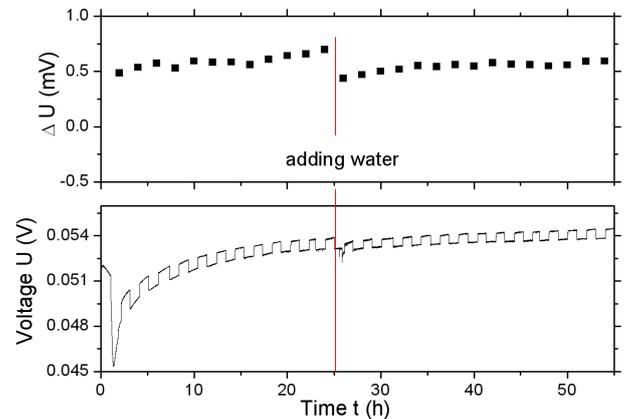


Fig. 5. Time evolution of the signal generated in the graphene based flow sensor. Lower part - measured signal, upper part - changes in amplitude caused by turning on and off the flow.

lems concerning the reproducibility of the results, we managed to obtain stable measurement conditions over a long period (Fig. 5). This is optimistic from the point of view of sensor applications, for which the sample is supposed to be continuously immersed in a flowing liquid.

Figure 5 shows the moment at which the measurement is disturbed by adding water (about 25 h). Moreover, we managed to obtain signals, which depend on the flow velocity.

Figure 6 shows the measured signals that show a linear dependence with the water flow rate. This result indicates that graphene can be successfully used in water flow sensors.

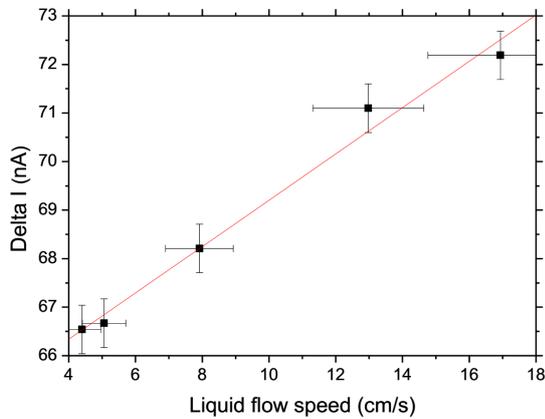


Fig. 6. Measured signals shows a linear dependence with the water flow speed in the range 4–18 cm/s.

#### 4. Conclusion

We studied voltage/current generation induced by a liquid flow in graphene. The best results were obtained on samples without metallization in the configuration with a Pt wire as a gate electrode and with an Ag/AgCl reference electrode. The flow-induced voltage/current is observed in deionized water aqueous NaCl or HCl solutions, as well as for ethanol. The observed signals are dominated by a component which is not dependent on the direction of liquid flow. This suggests that the main reason responsible for the observed voltage/current generation originates from the perturbation of the double layer that formed in the liquid close to the graphene surface. Further experimental work is needed to shed more light on the observed effects.

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