

# The Concept of the Leak Detecting Device for Liquid Sodium Using Graphene Oxide Sensors

Chungho Cho\*, Byung-Ho Kim, Jong-Man Kim, Youngil Cho, and Ji-Young Jeong  
Korea Atomic Energy Research Institute, 989-111 Daedeok-daero, Yuseong-gu, Daejeon, 305-353  
\*Corresponding author: chcho@kaeri.re.kr

## 1. Introduction

Liquid sodium easily reacts with oxygen or water in the air even at room temperature. Especially when high temperature liquid sodium leaks to the outside of the vessel or piping, it reacts with oxygen or moisture in the atmosphere and occurs a lot of fumes with flames. Therefore, it is necessary to detect the leakage of liquid sodium quickly by installing an appropriate leakage detection device at containers, piping, and equipment using liquid sodium.

Potential leak points of containers or piping systems using liquid sodium are where it is welded parts or high stresses and thermal fatigue are generated. In all processes including design, manufacture, quality assurance, operation and maintenance of containers or piping systems, rigorous inspections are taken to prevent leakage. But the possibility of leakage can't be entirely excluded. Therefore, if an event of liquid sodium leak occurs, it is very important to detect the leak early and cope with a sodium leak immediately to minimize leakage and prevent the spread of accidents.

In particular, in the case of double piping for transporting liquid sodium, it is not easy to install a conventional contact type sodium leak detector to detect the leakage of internal piping. So the analysis method for internal gases between the internal piping and the external protection piping is normally used to judge whether sodium leakage occurs.

Sodium Ionization Detector (SID), Radioactive Ionization Detector (RID), Laser Leak Detector (LLD) and Differential Pressure Detector (DPD) are known conventional methods for detecting sodium leakage through internal gas analysis. Disadvantages of sodium leakage detection methods through conventional internal gas analysis are summarized as follows.

(1) Frequent replacements of filaments or filters to maintain the leak detection function require a lot of time and cost.

(2) Since the large-sized and expensive devices or facilities such as individual radiation sources, shielding facilities, and lasers sources are need to setup the leak detection system, the spatial constraints and the installation cost of devices or facilities are burdensome.

This study aims to improve the previous liquid sodium leak detection methods by analyzing the internal gas.

This paper introduces the concept of sensing the leakage of liquid sodium using graphene oxide and a

simplified system with a plug-type liquid sodium leak detector.

## 2. The Concept of Graphene Oxide Sodium Sensor

### 2.1 Graphene

Graphene is a two-dimensional material with a continuous hexagonal honeycomb structure of carbon atoms. The single-layer graphene is a very thin material with a thickness of 3Å (0.1 nm) and is known as a semi-metallic material. When two-dimensional graphene are stacked in layers to form a three-dimensional structure, normally it is called Graphite.

Graphene is a physically and chemically very stable material and it can transport 100 times more current at room temperature than copper per unit area and can transport electrons 100 times faster than silicon.

Graphene with a thickness of 22 nm or less can flow 1,000 times more current than a copper wire, and the heat generation rate is only 1/10 of the copper wire [1, 2].

The strength of graphene is more than 200 times stronger than that of steel. Also, graphene does not lose intrinsic physical and electrical properties even when it folded completely because it has an excellent elasticity.

Particularly, one of the characteristics that should be noted here is that the material having the largest surface area relative to the same volume is graphene.

Due to these properties, the physical and chemical changes easily take place in the graphene surface through the reaction with gases. So the development of gas sensors using graphene has been actively conducted worldwide [3].

In addition, graphene has various bonding reactions ranging from very weak Van der Waals interactions to strong covalent bonds depending on the reactants.

The occurrence of this reactions can be easily confirmed by simply measuring the change in electrical resistance of graphene and it is considered to be an optimal method for determining the specific gas content in a confined space.

### 2.2 Graphene Gas Sensors

The sensitivity of the graphene gas sensor can be simply defined as  $\Delta R/R$  (R: inherent electrical resistance of the graphene before the gas reaction, and  $\Delta R$ : graphene electrical resistance change rate after the gas reaction). Because graphene exhibits an electric

behavior similar to that of a p-type semiconductor, when an electrons are lose through reactions with a specific gas, the electric resistance decreases. On the other hand, when electrons are obtained, the electric resistance increases.

For examples, when graphene reacts with gas such as NO<sub>2</sub> which deprive electrons from graphene, ΔR/R becomes negative, and when reacted with gas such as CO, NH<sub>3</sub> which supply electrons to graphene, ΔR/R has a positive value. Fig. 1 shows the experimental results of the electrical resistance change due to the reaction between graphene and various gases (NO<sub>2</sub>, CO, NH<sub>3</sub>, H<sub>2</sub>O) [4].

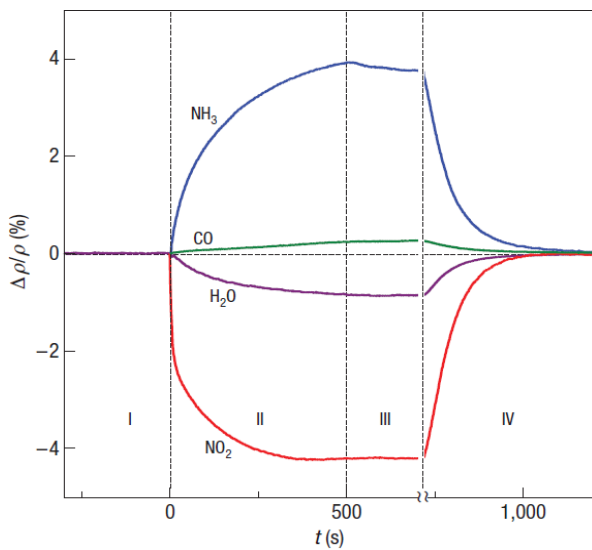


Fig. 1. The electrical resistance change of graphene due to the reaction between graphene and various gases [4].

### 2.3 The Concept of Graphene Oxide Sodium Sensor

It is known that graphene oxide is hydrophilic and is electrically insulated because it contain a large amount of the oxygen-containing functional group.

Fig. 2 shows the concept of structural bonding of graphene oxide containing a large amount of oxygen and hydroxide ions.

Pure graphene can be obtained by removing the oxygen-containing functional group from the graphene oxide. So various methods have been devised to remove oxygen-containing functional groups from the from the graphene oxide to get a pure graphene.

Under the given the fact that sodium reacts very easily with oxygen, sodium borohydride (NaBH<sub>4</sub>) has already been proposed to efficiently remove oxygen from graphene oxide for pure graphene production [5, 6].

Taking advantage of this fact, we propose a sensor with graphene oxide.

If the graphene oxide reacts with sodium vapor or sodium aerosol and we can detect changes in properties which occur in the process of changing from graphene oxide to pure graphene, the graphene oxide can be used as a sodium leakage detecting sensor.

Internal gases existing between the inner pipe and outer protective pipe are circulated to contact with a graphene sensor.

If the electrical resistance of the graphene oxide sensor changes like graphene gas sensor by reacting with the sodium aerosol or sodium vapor contained in the internal gases, it is possible to detect the leakage of the liquid sodium.

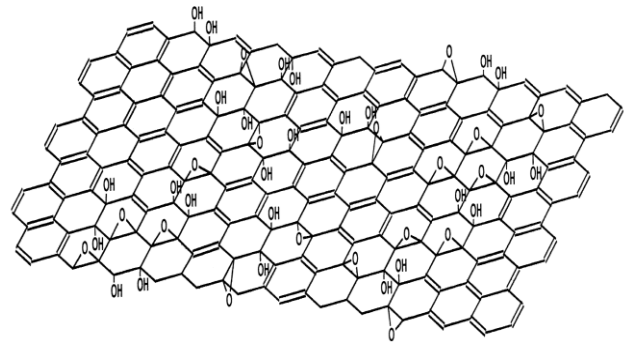
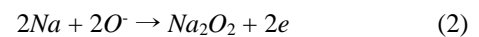
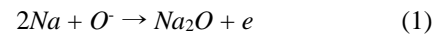


Fig. 2. The concept of structural bonding of graphene oxide

When a sodium aerosol or sodium vapor is contacted with graphene oxide, the following chemical reactions take place in the graphene oxide sensor.



Graphene oxide has negative electrical resistance changes because the sodium vapor or sodium aerosol deprived the oxygen-containing functional group from graphene oxide.

Fig. 3 shows the concept of the chemical reaction of oxygen-containing functional groups with sodium aerosol or sodium vapor.

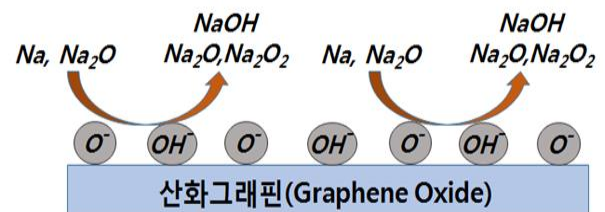


Fig. 3. The concept of the chemical reaction of oxygen-containing functional groups with sodium aerosol or sodium vapor

Fig. 4 shows a schematic diagram of a graphene oxide sodium sensor. The sensor takes the form of a plug with two electrodes to minimize installation space and increase installation convenience.

The upper part of the plug type sensor has a metal body part for binding the sensor to the leakage detection unit. The lower part of the plug type sensor is made of an insulator ( $\text{SiO}_2$  or something like a ceramic) to prevent the contact of two electrodes.

The upper ends of the electrode is used as a terminal for measuring the electrical resistance of the sensor, and a tube or a sheet of graphene oxide is bonded between the lower ends of the electrode.

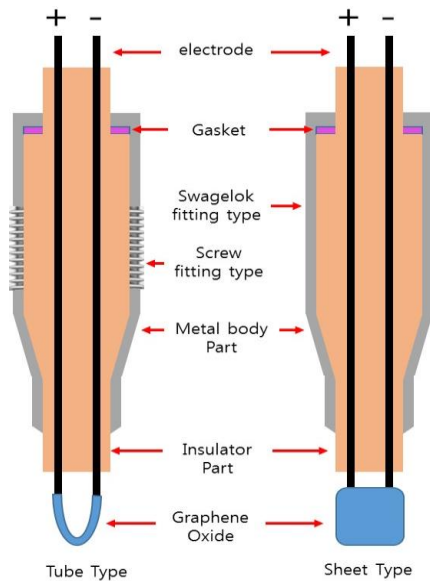


Fig. 4. Schematic diagram of a graphene oxide sodium sensor

### 3. Sodium leakage detection System for Dual Pipe

Fig. 5 shows the concept of a sodium leakage detection system for dual piping using graphene oxide sodium sensor.

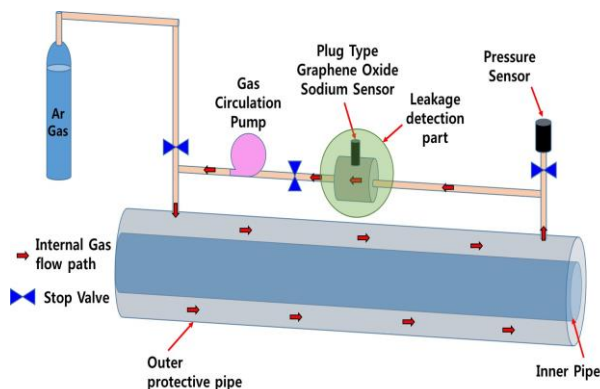


Fig. 5. The concept of a sodium leakage detection system for dual pipe

Between the inner piping and the outer protective piping fill with the inert gas to prevent fire due to the sodium leakage. Nitrogen gas can't be used as the inert gas because the oxygen-containing functional group of graphene oxide reacts with the nitrogen gas. So the Argon gas is used as the inert gas.

The internal gases are discharged from the rear end of the dual pipe by a gas circulating pump, and then gases those passes through a small container installed plug type graphene oxide sodium sensor come back again to the front end of the dual piping.

When liquid sodium is leaked from the inner piping, sodium aerosol or sodium vapor which are generated from leaking liquid sodium is present in the internal gases.

The internal gases are circulated and passed through a leak detection unit equipped with a plug type graphene oxide sodium sensor. Sodium sensor reacts with sodium aerosol or sodium vapor. A change in the electrical resistance of the sensor occurs and finally we can find out the sodium leakage of the inner piping of dual piping.

### 4. Conclusions

To improve the previous liquid sodium leak detection methods by analyzing the internal gas, the concept of liquid sodium leakage detecting device using graphene oxide is introduced.

The sensor takes the form of a plug type to minimize installation space and increase installation convenience.

The advantage of the plug type graphene oxide sodium sensor is that it does not needs expensive equipment and large facilities such as laser or radiation source shielding facility which are required by the previous liquid sodium leak detection system through the internal gas analysis.

The proposed graphene oxide sodium sensor in this study is still in the conceptual development study. Therefore, the performance, fabrication, applicability, and economics of the sensor applying this concept need to be verified through further research.

However, if the successful development of this sensor is achieved, this sensor is expected to be useful in the fields of sodium cooled fast reactor, nuclear fusion, lithium ion battery and energy saving system industry which handle alkaline liquid metal such as sodium or lithium.

### 5. Acknowledgement

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIP). (No. 2012M2A8A2025635)

## **REFERENCES**

- [1] Technology Status and R&D Trends of Next Generation New Material Graphene, Ha Yeon Co., 2011.
- [2] KISTI, Graphene, the new material of dreams, won the 2010 Nobel Prize, A Scientific flavor, No. 1243, 2010.11.01.
- [3] Yeo S. M., et al., Development of high sensitivity graphene gas sensor through ion beam irradiation, KAERI/RR-4168/2016.
- [4] F. Schedin, et al., Detection of Individual Gas Molecules Adsorbed on Graphene, Nature Materials, Vol. 6. pp. 652, 2007.
- [5] Shin H J, et al., Efficient reduction of Graphite Oxide by Sodium borohydride and its effect on electrical conductance, Advanced Functional Materials, 2009.
- [6] Zhen-zhen Yang, et al., A Simple Method for the reduction of Graphene Oxide by Sodium borohydride with  $\text{CaCl}_2$  as a Catalyst, New Carbon Materials, Vol. 30, Issue 1, 2015.
- [7] Heyong He, et al., A Structural model for Graphite Oxide, Chemical Physics Letters, 1998.