**Preparation of graphene oxide/silica composite membranes and their application for nanofiltration**

Kojiro Ueda1, Masanobu Nakata1, Hideki Yamamoto1, Sadao Araki1\*

*1: Kansai University, Department of Chemical, Energy and Environmental Engineering, 3-3-35 Yamate-cho, Suita-shi, Osaka 564-8680 Japan*

*\*Corresponding author: araki\_sa@kansai-u.ac.jp*

**Highlights**

* Graphene oxide/Silica composite (GO/SiO2) membrane was prepared by using hydrolysis and polycondensation reaction of tetraethoxysilane.
* GO and GO/SiO2 membranes showed high rejection of rose bengal.
* GO/SiO2 membrane showed high adhesion properties compared to GO membrane.

**1. Introduction**

Graphene oxide (GO) multi-layered membranes have high permeability and good separation properties. However, since it is formed just by laminating GO on a support, GO layers easily peel off in plug flow type modules and the performance is remarkably decreased. In this study, to solve this issue of GO membranes, graphene oxide/Silica composite (GO/SiO2) membranes were focused. It is expected that silanol groups react with hydroxyl groups of GO. Therefore, it is thought that GO/SiO2 membranes have high durability compared with GO and polymer composite membranes. We prepared the GO/SiO2 membranes and their nanofiltration properties was confirmed. In addition, the effect of composite of GO and silica on their adhesion was evaluated.

**2. Methods**

2.1 Preparation of GO membrane

GO suspension was synthesized using a modified Hummer׳s method and its concentration was 0.05 g/L [1]. [Yttrium](https://www.sciencedirect.com/topics/materials-science/yttrium) stabilised [zirconia](https://www.sciencedirect.com/topics/materials-science/zirconia) (YSZ) hollow fiber support with dead-end structure was soaked in the suspension. GO layers were deposited on the support by vacuuming inside of the support for 30 minutes. Then, the membrane was dried in air for 2 days.

2.2 Preparation of GO/SiO2 membrane

0.1 mol of tetraethoxysilane (TEOS) as a silica source was added to 50 ml of ethanol. As an acid catalyst, 15 ml of 1 mol/ml nitric acid was added at a dropping rate of 1 ml/min. By stirring for 3 h, silica sol was prepared. GO was dispersed in the silica sol at a concentration of 0.05 g/L, and GO/SiO2 membrane was prepared by the same procedure as the GO membrane.

2.3 Nanofiltration test

GO and GO/SiO2 membranes were tested by connecting them in a pressure vessel filled with 35 µM rose bengal aqueous solution. The filtration tests were conducted at 5 bar. The permeation flux was calculated from the weight of the permeate. The dye concentration of the permeate was determined by using a UV-spectrometer (ASUV-6300PC, Shimadzu). The rejection rate calculated from the concentration ratio of permeate to feed solution. In addition, to confirm the adhesion of GO and GO/SiO2 membranes, the pressure of 2 bar was added from inside of the fiber. Then, their nanofiltration properties were confirmed by 35 µM rose bengal solution.

**3. Results and discussion**

Table 1 shows nanofiltration results of GO and GO/SiO2 membranes. The permeance of GO/SiO2 membrane was reduced to one-third of the GO membrane. It is thought that SiO2 inhibited permeation of water molecules. The rejections of both GO and GO/SiO2 membranes was almost 100%. Table 2 shows nanofiltration results of GO and GO/SiO2 membranes after the applying counter pressure. The rejection of GO membrane was reduced to 0% because GO layers were peeled off. On the other hand, GO/SiO2 membrane kept a high rejection rate. From the above results, compared to the GO membrane, the adhesion properties of the GO/SiO2 membrane were improved by the siloxane network.







**4. Conclusions**

GO/SiO2 membrane was prepared to improve the adhesion properties of GO membranes. GO/SiO2 membrane showed a high rejection of rose bengal, which is similar with the GO membrane. After the addition of counter pressure, the rejection of rose bengal for GO membrane was dramatically reduced. On the other hand, GO/SiO2 membrane kept the rejection after the applying counter pressure.

**References**

1. Liu. Hong. et al, J. Ferroelectrics. 528 (2017) 15-21