

## Supporting Information

# Selective Protein Transport through Ultra-thin Suspended Reduced Graphene Oxide Nanopores

*Dae-Sik Lee<sup>a</sup>, Seokhan Park<sup>b</sup>, Yong Duk Han<sup>c</sup>, Jae Eun Lee<sup>d</sup>, Hu Young Jeong<sup>e</sup>, Hyun C. Yoon<sup>c</sup>, Moon Yeon Jung<sup>a</sup>, Sang Ouk. Kim<sup>b\*</sup>, Sung-Yool Choi<sup>a, d\*</sup>*

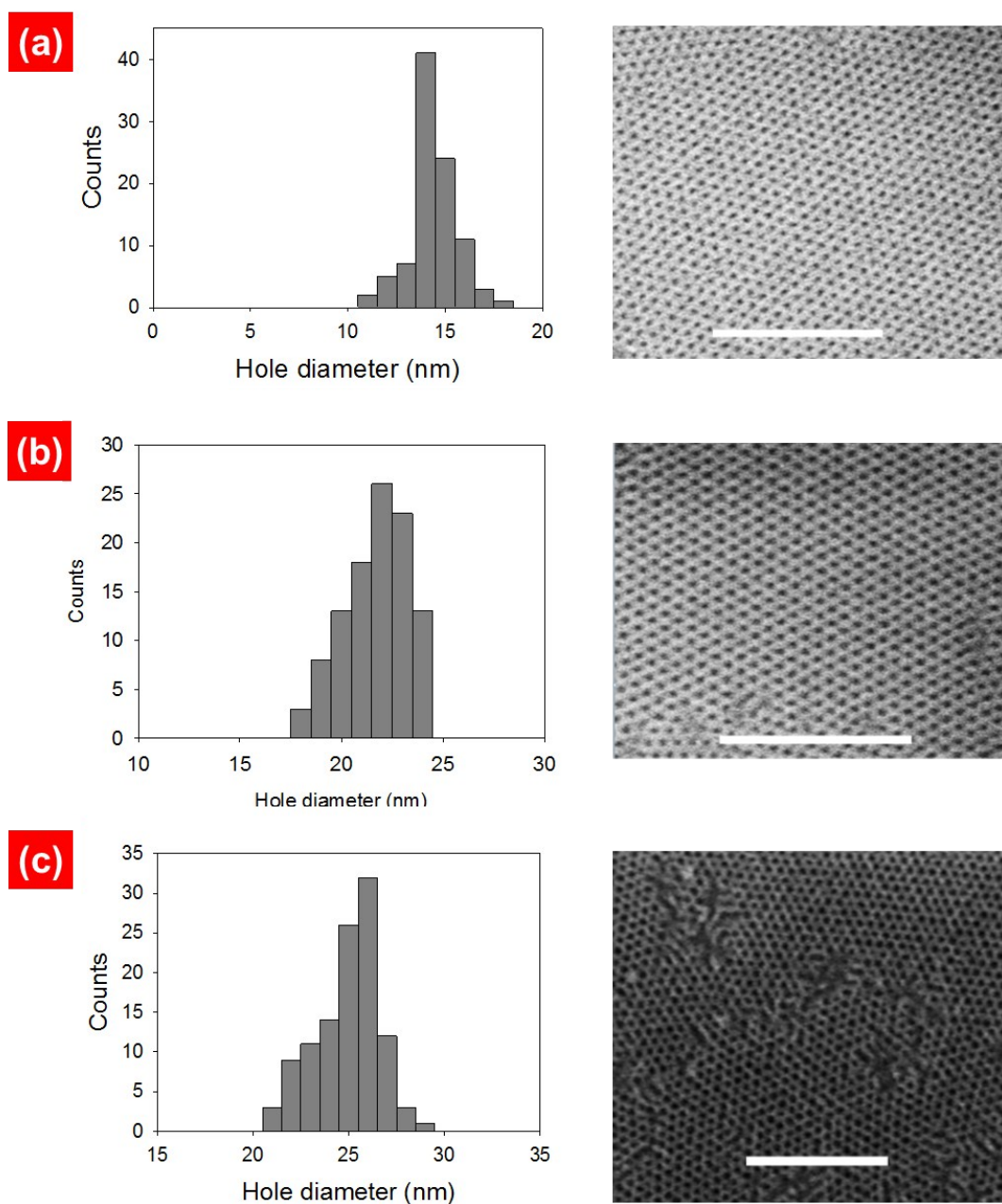
<sup>a</sup> Electronics and Telecommunications Research Institute (ETRI), Daejeon, Republic of Korea.

<sup>b</sup> Department of Materials Science and Engineering, KAIST, Daejeon 34141, Republic of Korea.

<sup>c</sup> Department of Molecular Science and Technology, Ajou University, Suwon 16499, Republic of Korea.

<sup>d</sup> School of Electrical Engineering, Graphene Research Center, KAIST, Daejeon 34141, Republic of Korea.

<sup>e</sup> UNIST Central Research Facilities(UCRF), UNIST, 50 UNIST-gil, Ulsan 44919, Republic of Korea



**Figure S1.** Pictures with illustrations along with the SEM images employing the spin casting block copolymer as an etch mask and a reactive ion etching protocol in order to form nanopores precisely and uniformly. The distribution of minimal nanopore diameters on the upper side of the membrane and pore size data were extracted from 320 and 360 pores which were selected randomly from numerous pores shown in the SEM images, changing etching time from 100 s (a), 106 s (b) and 112 s (c). (Scale bar indicates 500 nm)

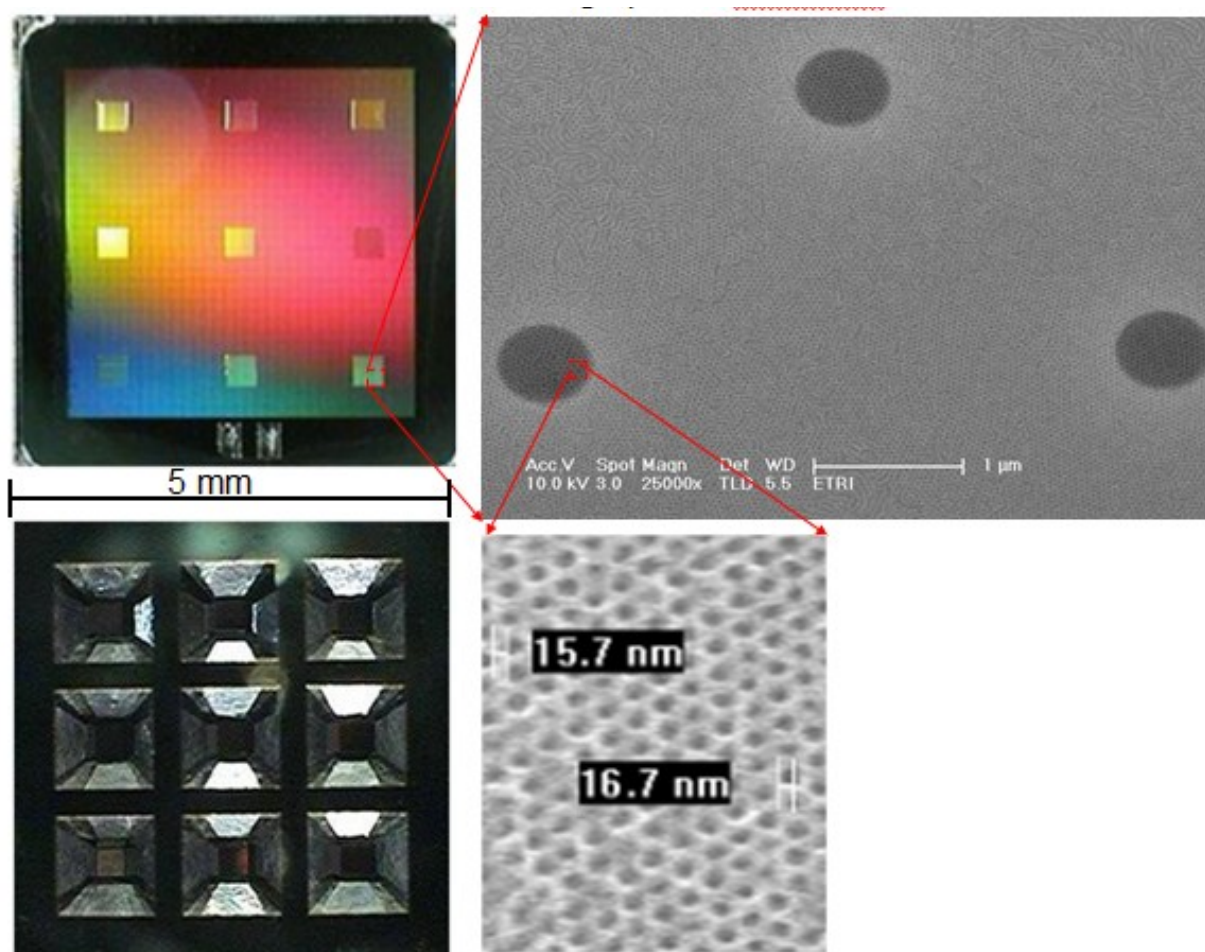
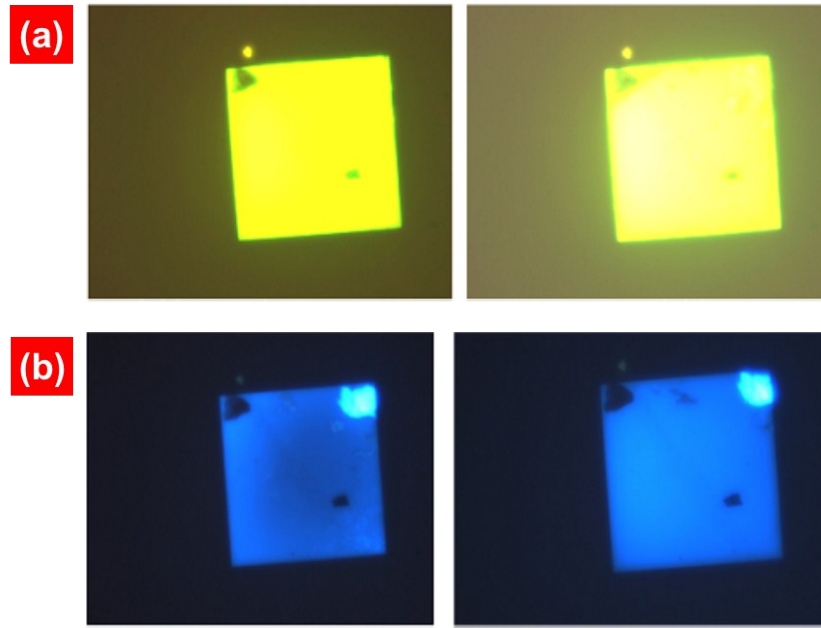


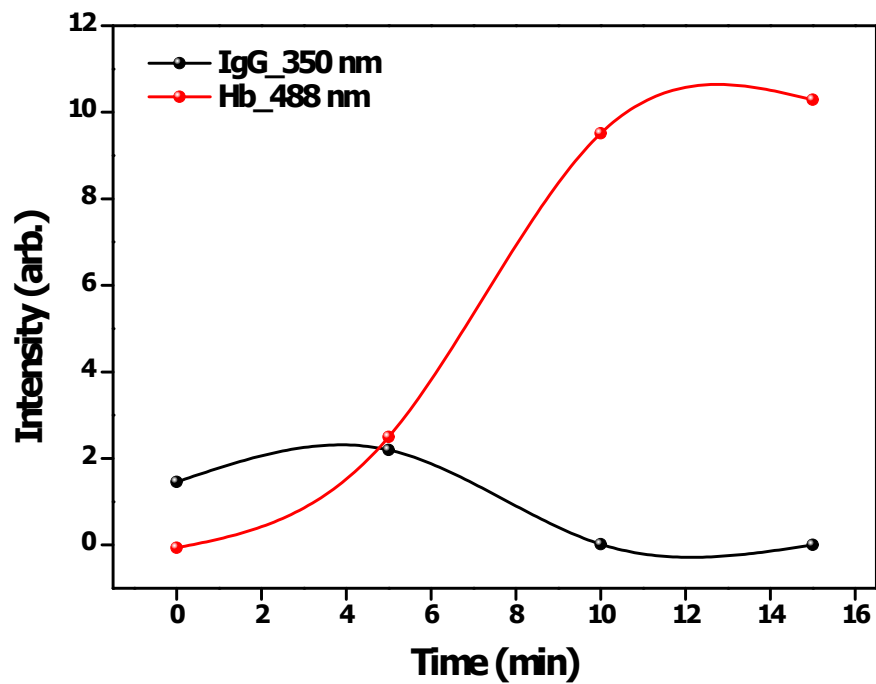
Figure S2. Photographs of the nanosieve device; top view and bottom view, and magnified SEM photographs showing the reduced graphene on the membranes



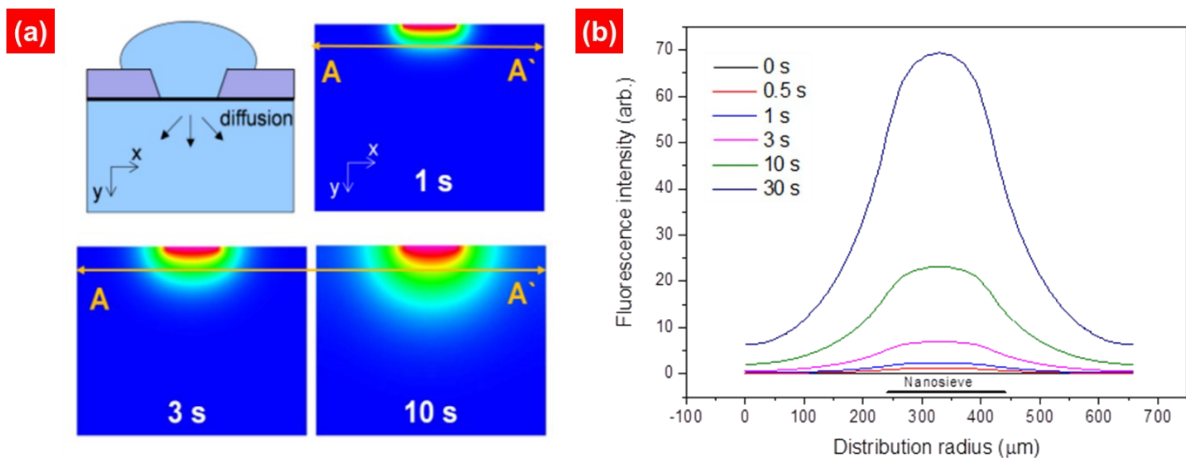
**Figure S3.** Fluorescence photographs showing selective penetration of mixture of Hemoglobin (Hb) (a) and Immunoglobulin G (IgG) (b) at separation-starting time and after 10 min in the nanosieve with SiN membrane, IgG is tagged with Alexa Fluor® 350 and Hb with Alexa Fluor® 488.

In Figure S3, the fluorescence of the labeled IgG at the center part of the supporting membrane clearly increases from 0 to 10 min, even though the increase in the signal is, generally, small in comparison with the fluorescence of the Alexa Fluor® 488-labeled Hb. If there were to be any ruptures or breakages in the center of the membrane, there would be a drastic increase in the amount of IgG with Alexa Fluor® 350 at the observation site, 50  $\mu\text{m}$  from the edge of the supporting membrane (200  $\mu\text{m}$  x 200  $\mu\text{m}$ ). On the other hand, the fluorescence of the labeled IgG decreases over time, as shown in Figure S4. This figure shows the intensity increase of the Hb with the Alexa Fluor® 488 dye and the intensity decrease of the IgG with the Alexa Fluor® 350 over time.

We think that the fluorescence increase of the labeled IgG at the center of the supporting membrane is caused by existence of the optically transparent SiN membranes, in which the exciting light usually gives rise to signal interference, such as scattering or reflections, as observed by the CCD detectors. When the noise level of the signals is low, in particular, the effects of the transparent membrane would increase.



**Figure S4.** Fluorescence intensity as a function of penetration time using a mixture of Hb and IgG proteins in the nanosieve with SiN membrane. The signals were measured at a 50  $\mu\text{m}$ -away from the edge of the supporting membrane (200  $\mu\text{m}$  x 200  $\mu\text{m}$ ).



**Figure S5.** Simulation result of the diffused Alexa Fluor® 488 in water. 2-D images of diffused Alexa Fluor® 488 after 1 s, 3 s, 10, and 30 s (a) and the distribution of the permeated Alexa Fluor® 488 in regards to the permeation time along line from A to A' (b).

**Supporting\_video 1.** Fluorescence images of the Alexa Fluor® 488-labeled hemoglobin (100 µg/ml) as a function of the permeation time using the a mixture of Hb and IgG proteins permeated through the graphene nanosieve

**Supporting\_video 2.** Fluorescence images of the Alexa Fluor® 350-labeled IgG (100 µg/ml) as a function of the permeation time using the a mixture of Hb and IgG proteins permeated through the graphene nanosieve device

**Supporting\_video 3** Permeation images of the trapped air bubble among the Alexa Fluor® 350-labeled IgG (100 µg/ml) using the a mixture of Hb and IgG proteins permeated through the graphene nanosieve device as a function of the permeation time