Deterministic Structuring of 2D Materials at the Nanoscale: Towards a Large-scale Nano-sieve for Gas Separation

First of all, I would like to express my great gratitude and appreciation to the Åforsk Foundation for the invaluable support through the Young Researcher grant. The support from the Foundation has helped me tremendously to realize the research ideas and helped the project get off the ground. Personally, the grant is very meaningful to me as an early-career researcher because it makes me more confident in pursuing and developing this line of research. Thank you very much for your support!

Through this funding, we have been able to achieve most objectives in the initial proposal. With the assistance from my master student, Hendrik Bruce, we have developed thoroughly the fabrication process of the self-supporting masks with the through-hole nanopatterns using the micro-nano fabrication facility at Uppsala University (Myfab Uppsala). The advantage of this fabrication process is that it provides high customizability in the structure of the patterns due to the flexibility of the electron beam writing process. Fig. 1 shows the simplified process flow and the fabricated through-hole nanopatterns of different sizes.

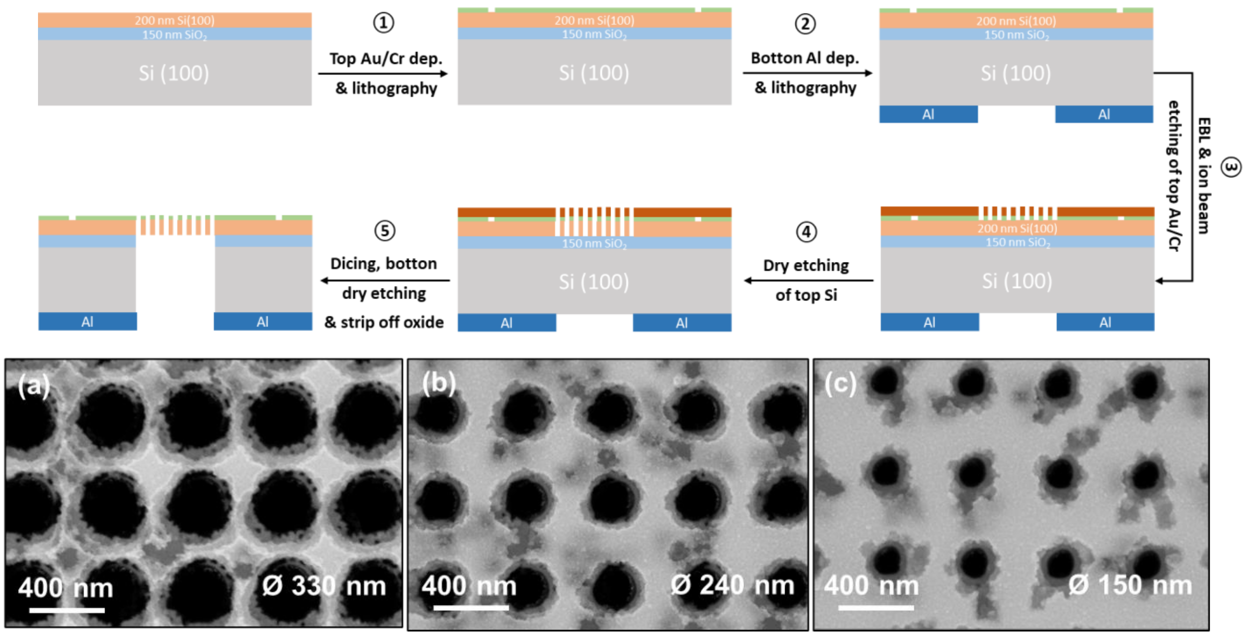

Fig. 1: The fabrication process of the self-supporting mask and the SEM images of the masks with the through-hole nanopatterns.

Using the fabricated mask and the graphene samples purchased from Graphenea, we have conducted successfully the structuring process of the self-supporting graphene samples. Fig. 2 shows the self-supporting graphene membranes after being structured with ion irradiation through the self-supporting mask. The patterns induced on graphene are determined by the patterns of the mask. The pore size achieved in this experiment is down to 15 nm, closely approaching the spatial resolution of the focused electron beam and the ion beam techniques. The focused beam methods, however, cannot be applied over large areas and hence have very limited scalability. Our method can be applied to large-area membranes, which is more suitable for future applications. Furthermore, our method is contactless, and does not use any chemical and mechanical processes. Hence, it is much more applicable for the delicate self-supporting films.

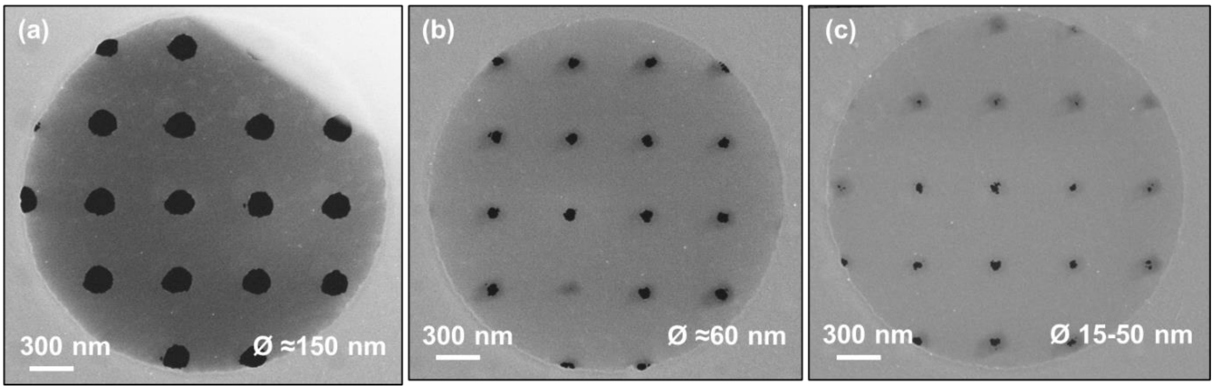

Fig. 2: Self-supporting graphene membrane after structured using the through-hole nanopattern mask and ion irradiation.

In addition to the structuring capabilities of the self-supporting graphene and other 2-dimensional materials, we also found that the method makes the membranes cleaner from the original contaminations which is a current severe issue for large-area graphene grown by chemical vapor deposition. The results show that our methods have multiple advantages, such as the large-scale structuring, the cleaning effect, the suitability, and the simplicity of the structuring process. These results have been published in a prominent journal for 2D materials research (2D Materials, impact factor 6.2): **Tran, T. T., et al. (2023). "A contactless single-step process for simultaneous nanoscale patterning and cleaning of large-area graphene." 2D Materials 10(2): 025017**. Because of this project, I was invited to present the results at the German Physical Society in Dresden in March 2023. Scientifically, through the employment of this unique experimental setup we have found a strong correlation between the level of the original mobile contaminants and the stability of the graphene lattice. The result suggests that atomic defects generated within the irradiated areas can diffuse into the non-irradiated area at room temperature and a certain portion of defects are annihilated by the mobile contaminants. This effect is observed by various electron microscopy and diffraction methods available at our disposal. **The second manuscript related to the graphene project will be prepared by the end of this year.**

For testing the functionality of the structured graphene for filtering applications, we have started to construct the filtering setup as shown in Fig. 3. Most parts of the setup, in particularly the two diffusion pumps, are recycles from other used equipment. Other components, such as the filter valve for mounting the filtering membranes and the gas system, will be completed early next year with the support from an engineer of the Tandem Laboratory, Bart Royeaerd.

Fig. 3: Setup for testing the filtering function of the graphene membrane (under construction)

Another outcome of the graphene project is that we have developed a fabrication process for self-supporting germanium (Ge) membranes. Although the Ge membranes are another line of research we are pursuing, the fabrication process for the Ge membranes is based on the process for the nanopattern mask. **We have prepared and submitted a manuscript describing the fabrication process of the Ge membrane to the journal Material Science for Semiconductor Processing**.

In summary, the funding from Åforsk has been tremendously impactful in realizing the research ideas. The results of the project have directly led to one peer-reviewed publication, another under preparation, and indirectly to another publication under review. Relying on the groundwork of these results, I am conceptualizing this experimental methodology into a versatile approach for structuring, functionalizing, and engineering of atomic defects in 2D materials. Such materials with the desired modification are essential for a number of emerging technologies, such as nanoelectronics, single-atom catalysis, single-photon emission and nanofiltration.

The following table shows how the funding has been used within our research activities:

Purposes	**Amount, SEK**
Researcher salary	294.836
Engineering and workshop service	101.178
Distributed indirect cost	106.342
Distribution of internal rent	26.159
Conference expenses	12.424
IT fee	3972

Total	**544.911**

Thank you very much for your invaluable support!

With best regards,

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