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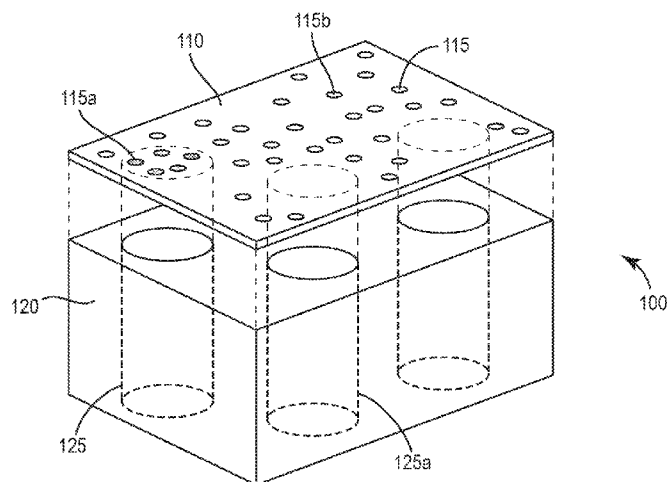


FIG. 1

(57) Abstract: A two-dimensional membrane layered structure may include a support substrate layer having a plurality of substrate passages configured to allow fluid to flow therethrough, a two-dimensional membrane layer disposed on an upper surface of the support substrate layer, and a plurality of flow passages disposed between the support substrate layer and the two-dimensional membrane layer. The two-dimensional membrane layer may have a plurality of pores configured to allow fluid to flow therethrough. The plurality of pores may comprise a first portion of pores that do not overlap with the plurality of substrate passages and a second portion of pores that do not overlap with the plurality of substrate passages. The plurality of flow passages may be configured to allow fluid to flow through the second portion of pores to the plurality of substrate passages.



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TWO-DIMENSIONAL MEMBRANE STRUCTURES HAVING FLOW PASSAGES**CROSS-REFERENCE TO RELATED PATENT APPLICATIONS**

[0001] The present application claims priority to co-pending U.S. Application No. 15/099,420 filed April 14, 2016, titled “TWO-DIMENSIONAL MEMBRANE STRUCTURES HAVING FLOW PASSAGES,” which is incorporated by reference herein in its entirety.

BACKGROUND

[0002] Two-dimensional membranes are materials with a thickness on an atomic scale. Potential applications of two-dimensional membranes are widespread, including their use in filtration, barrier, and separation devices. Due to their atomic-level thickness, however, two-dimensional membranes often need to be disposed onto a support substrate to provide the needed mechanical support for its intended use. In addition, the support substrate may provide enhanced biocompatibility and/or easier device integration to the membrane structure.

[0003] When a two-dimensional membrane is applied to a support substrate, a large fraction of the pores present in the two-dimensional membrane do not overlap with the passages present in the support substrate. Thus, fluid may only flow through those pores that overlap with the support substrate passages. Those pores that do not overlap with the support substrate passages are not utilized, resulting in low overall pore utilization in the membrane. In many cases, the overall pore utilization for these structures can be as low as 5%, leading to a lower overall fluid flow through the two-dimensional membrane structure.

[0004] To increase flow through the two-dimensional membrane structure and to increase overall pore utilization, one method may be to increase the density of pores in the two-dimensional membrane such that the amount of pores that overlap with the support substrate channels is increased. However, increasing the density of pores in the membrane may add to the mechanical strain on the two-dimensional membrane and may make transferability of large-scale membranes (e.g., $> 1 \text{ cm}^2$) to the support substrate difficult.

SUMMARY

[0005] In some embodiments, a two-dimensional membrane layered structure may include a support substrate layer having a plurality of substrate passages configured to allow fluid to flow therethrough, a two-dimensional membrane layer disposed on an upper surface of the support substrate layer, and a plurality of flow passages disposed between the support substrate layer and the two-dimensional membrane layer. The two-dimensional membrane layer may have a plurality of pores configured to allow fluid to flow therethrough. The plurality of pores may comprise a first portion of pores that overlap with the plurality of substrate passages and a second portion of pores that do not overlap with the plurality of substrate passages. The plurality of flow passages may be configured to allow fluid to flow through the second portion of pores to the plurality of substrate passages.

[0006] In some embodiments, the two-dimensional membrane layered structure may comprise a plurality of interlayer supports disposed on an upper surface of the support substrate layer. The plurality of interlayer supports may form the plurality of flow passages.

[0007] In some embodiments, the two-dimensional membrane layered structure may comprise a plurality of grooves formed on an upper surface of the support substrate layer. The plurality of grooves may form the plurality of flow passages.

[0008] In some embodiments, the two-dimensional membrane layered structure may comprise a plurality of interlayer supports disposed on an upper surface of the support substrate layer and a plurality of grooves formed on an upper surface of the support substrate layer. The plurality of interlayer supports and the plurality of grooves may form the plurality of flow passages.

[0009] In some embodiments, a two-dimensional membrane layered structure may include a support substrate layer having a plurality of substrate passages configured to allow fluid to flow therethrough, a two-dimensional membrane layer disposed on an upper surface of the support substrate layer, and a plurality of interlayer supports disposed on the upper surface of the support substrate layer. The two-dimensional membrane layer may have a plurality of pores configured to allow fluid to flow therethrough. The plurality of pores may comprise a first portion of pores that overlap with the plurality of substrate passages and a second portion of pores that do not overlap with the plurality of substrate passages. The

plurality of interlayer supports may be configured to form a plurality of flow passages that allow fluid to flow through the second portion of pores to the plurality of substrate passages.

[0010] In some embodiments, the plurality of interlayer supports may comprise carbon nanotubes.

[0011] In some embodiments, the plurality of interlayer supports may comprise electrospun fibers.

[0012] In some embodiments, the plurality of interlayer supports may be disposed on an area of the upper surface of the support substrate layer ranging from about 5% to about 50% of the total area of the upper surface of the support substrate layer.

[0013] In some embodiments, the plurality of interlayers supports may be functionalized.

[0014] In some embodiments, the plurality of interlayer supports may be disposed on an area of the upper surface of the support substrate layer ranging from about 40% to about 45% of the total area of the upper surface of the support substrate layer.

[0015] In some embodiments, a two-dimensional membrane layered structure may comprise a support substrate layer having a plurality of substrate passages configured to allow fluid to flow therethrough, a two-dimensional membrane layer disposed on an upper surface of the support substrate layer, and a plurality of grooves formed on the upper surface of the support substrate layer. The two-dimensional membrane layer may have a plurality of pores configured to allow fluid to flow therethrough. The plurality of pores may comprise a first portion of pores that overlap with the plurality of substrate passages and a second portion of pores that do not overlap with the plurality of substrate passages. The plurality of grooves may be configured to form a plurality of flow channels that allow permeate to flow through the second portion of pores to the plurality of substrate channels.

[0016] In some embodiments, the plurality of grooves may be formed on the upper surface of the support substrate layer in a lattice pattern.

[0017] In some embodiments, the plurality of grooves may comprise a width of about 10 nanometers to about 1000 nanometers.

[0018] In some embodiments, the plurality of grooves may be functionalized.

[0019] In some embodiments, a method of increasing pore utilization in a two-dimensional membrane layered structure may comprise providing a substrate support layer having a plurality of substrate passages configured to allow fluid to flow therethrough, disposing a plurality of interlayer supports on an upper surface of the substrate support layer, and disposing a two-dimensional membrane layer having a plurality of pores configured to allow fluid to flow therethrough on the upper surface of the substrate support layer having the plurality of interlayer supports. The plurality of pores may comprise a first portion of pores that overlap with the plurality of substrate passages and a second portion of pores that do not overlap with the plurality of substrate passages. The plurality of interlayer supports may form a plurality of flow passages. The plurality of flow passages may be configured to allow fluid to flow through the second portion of pores to the plurality of substrate passages.

[0020] In some embodiments, the plurality of interlayer supports may comprise carbon nanotubes and the carbon nanotubes may be disposed on the upper surface of the support substrate layer via spray-coating.

[0021] In some embodiments, a method for increasing pore utilization in a two-dimensional membrane layered structure may comprise providing a substrate support layer having a plurality of substrate passages configured to allow fluid to flow therethrough, forming a plurality of grooves on an upper surface of the substrate support layer, and disposing a two-dimensional membrane layer having a plurality of pores configured to allow fluid to flow therethrough on the upper surface of the substrate support layer having the plurality of grooves. The plurality of pores may comprise a first portion of pores that overlap with the plurality of substrate passages and a second portion of pores that do not overlap with the plurality of substrate passages. The plurality of grooves may form a plurality of flow passages. The plurality of flow passages may be configured to allow fluid to flow through the second portion of pores to the plurality of substrate passages.

[0022] In some embodiments, the plurality of grooves may be formed in a lattice pattern.

[0023] In some embodiments, a two-dimensional membrane layered structure may comprise a support substrate layer having a plurality of substrate passages configured to allow fluid to flow therethrough, and a two-dimensional membrane layer disposed on an

upper surface of the support substrate layer. The two-dimensional membrane layer may have a plurality of pores configured to allow fluid to flow therethrough. The plurality of pores may comprise a first portion of pores that overlap with the plurality of substrate passages and a second portion of pores that do not overlap with the plurality of substrate passages. The two-dimensional membrane layered structure may further comprise a flow passage means for allowing fluid to flow through the second portion of pores to the plurality of substrate passages to increase overall pore utilization of the two-dimensional membrane layer.

BRIEF DESCRIPTION OF THE DRAWINGS

[0024] FIG. 1 is a perspective view of a two-dimensional membrane layered structure according to one embodiment.

[0025] FIG. 2 is a perspective of the two-dimensional membrane layered structure of FIG. 1 having fluid flow passages.

[0026] FIG. 3 is a cross-sectional view of the two-dimensional membrane layered structure having the flow passages of FIG. 2 according to a first embodiment.

[0027] FIG. 4 is a cross-sectional view of the two-dimensional membrane layered structure having the flow passages of FIG. 2 according to a second embodiment.

[0028] FIG. 5 is a scanning electron microscope (SEM) micrograph of a support substrate layer having interlayer supports to form fluid flow passages.

[0029] FIG. 6 is a detailed SEM micrograph of the support substrate layer of FIG. 5.

DETAILED DESCRIPTION

[0030] Some embodiments relate to a two-dimensional membrane layered structure having a plurality of flow passages formed between a two-dimensional membrane layer and a support substrate layer. The flow passages provide an increase in pore utilization by allowing fluid to flow through pores in the two-dimensional membrane layer that do not overlap with passages present in the support substrate. With the flow passages, fluid may flow from the non-overlapping pores through the flow passages to a nearby support substrate passage. In some embodiments, the flow passages may be formed by interlayer supports laterally

disposed on an upper surface of the support substrate layer and/or by grooves laterally formed on the upper surface of the support substrate layer. In addition to increasing overall pore utilization of the membrane layer and total flow through the membrane structure, the flow passages may also provide sufficient support to the two-dimensional membrane without adding undesirable strain to the membrane layer.

[0031] FIG. 1 shows a perspective view of a two-dimensional membrane layered structure 100. The two-dimensional membrane layered structure 100 may include a two-dimensional membrane layer 110 having a plurality of pores 115, and a support substrate layer 120 having a plurality of substrate passages 125 that extend along a thickness of the support substrate layer 120.

[0032] The two-dimensional membrane layer 110 may be a single-layer two-dimensional material or a stacked two-dimensional material. Most generally, a single-layer two dimensional material is atomically thin, having an extended planar structure and a thickness on the nanometer scale. Single-layer two-dimensional materials generally exhibit strong in-plane chemical bonding relative to the weak coupling present between layers when such layers are stacked. Examples of single-layer two-dimensional materials include metal chalcogenides (e.g., transition metal dichalcogenides), transition metal oxides, boron nitride (e.g., hexagonal boron nitride), graphene, silicone, germanene, carbon nanomembranes (CNM), and molybdenum disulfide. Stacked two-dimensional materials may include a few layers (e.g., about 20 or less) of a single-layer two-dimensional material or various combinations of single-layer two-dimensional materials. In some embodiments, the two-dimensional membrane layer 110 may be a graphene or graphene-based two-dimensional material as a single-layer two-dimensional material or a stacked two-dimensional material.

[0033] The support substrate layer 120 may include any appropriate planar-type substrate. In some embodiments, the support substrate layer 120 may be made from ceramic porous materials, such as silica, silicon, silicon nitride, and combinations thereof. In some embodiments, the support substrate layer 120 may be made from a polymer material, such as track-etched polymers, expanded polymers, patterned polymers, non-woven polymers, and combinations thereof. The support substrate layer 120 may include a polymer selected from the group consisting of polysulfones, polyurethane, polymethylmethacrylate (PMMA), polyethylene glycol (PEG), polylactic-co-glycolic acid (PLGA), PLA, PGA, polyamides

(such as nylon-6,6, supramid and nylamid), polyimides, polypropylene, polyethersulfones (PES), polyvinylidene fluoride (PVDF), cellulose acetate, polyethylene, polypropylene, polycarbonate, polytetrafluoroethylene (PTFE) (such as Teflon), polyvinylchloride (PVC), polyether ether ketone (PEEK), mixtures and block co-polymers of any of these, and combinations and/or mixtures thereof. In some embodiments, the polymers may be biocompatible, bioinert and/or medical grade materials. In some embodiments, the support substrate layer 120 may be a track-etched polycarbonate (TEPC) membrane. In other embodiments, the support substrate layer 120 may be a membrane formed by silicon nitride, track-etched polyimide, track-etched polyester, track-etched SiN, nanoporous silicon, nanoporous silicon nitride, an electrospun membrane, and/or a PVDF membrane. In some embodiments, the substrate passages 125 may be formed randomly or in a patterned manner.

[0034] As shown in FIG. 1, when disposing the two-dimensional membrane layer 110 on the support substrate 120, only a certain percentage of pores 115 overlap with the substrate passages 125. For example, a first portion of pores 115a overlap with, or are in fluid communication with, the support substrate passages 125. The first portion of pores 115a contributes to overall pore utilization because they allow fluid to flow through to the substrate passages 125. On the other hand, a second portion of pores 115b do not overlap with, or fail to be in fluid communication with, the support substrate passages 125. Thus, the second portion of pores 115b does not contribute to overall pore utilization because fluid cannot flow from these pores into the support substrate passages 125. Moreover, as further shown in FIG. 1, in some cases, a support substrate passage 125a may fail to be in fluid communication with any of the pores 115. Thus, this blocked support substrate passage 125a receives no fluid flow, thereby decreasing the overall utilization of the support substrate passages 125.

[0035] FIG. 2 illustrates a perspective view of the two-dimensional membrane layered structure 100 having a plurality of flow passages 155 disposed between the two-dimensional membrane layer 110 and the support substrate layer 120. As shown in FIG. 2, the flow passages 155 provide a means for the second portion of pores 115b to be utilized by providing a flow path for fluid to flow from a respective pore 115b to a nearby substrate passage 125. In some embodiments, the flow passages 155 may also provide a flow path from the second portion of pores 115b to the blocked support substrate passage 125a such that fluid may flow through the blocked support substrate passage 125a. Thus, the integration of the flow passages 155 provide for an increase in the overall pore utilization of the two-

dimensional membrane layer 110 and a means for increased fluid flow through the support substrate layer 120.

[0036] FIG. 3 shows a cross-sectional view of the two-dimensional membrane layered structure 100 having flow passages 155 formed therein in accordance with a first embodiment. As shown in FIG. 3, a plurality of interlayer supports 150 may be disposed laterally on an upper surface 126 of the support substrate layer 120. The two-dimensional membrane layer 110 is then disposed onto the upper surface 126 of the support substrate layer 120 having the interlayer supports 150 disposed thereon. When layered, the interlayer supports 150 support the two-dimensional membrane layer 110 while pushing the two-dimensional membrane layer 110 upward from the support substrate layer 120 to form the flow passages 155 that allow fluid to flow through the pores 115b.

[0037] The interlayer supports 150 may take numerous forms. For example, in some embodiments, the interlayer supports 150 may be carbon nanotubes. For example, FIGS. 5 and 6 show interlayer supports in the form of carbon nanotubes spray-coated onto an upper surface of a support substrate layer in the form of a track-etched polyimide layer before the two-dimensional membrane layer is disposed onto the support substrate layer. As shown the figures, the carbon nanotubes are disposed onto the upper surface of the support substrate layer so as to extend laterally across the upper surface to provide access for fluid flow to the substrate passages. In some embodiments, the carbon nanotubes may be single-walled or multi-walled.

[0038] In other embodiments, the interlayer supports 150 may be electrospun fibers. In yet other embodiments, the interlayer supports 150 may be nanorods, nanoparticles (e.g., oxide nanoparticles, octadecyltrichlorosilane nanoparticles), fullerenes, collagen, keratin, aromatic amino acids, polyethylene glycol, lithium niobate particles, decorated nano-dots, nanowires, nanostrands, lacey carbon material, proteins, polymers (e.g., hygroscopic polymer, thin polymer, amorphous polymer), hydrogels, self-assembled monolayers, allotropes, nanocrystals of 4-dimethylamino-N-methyl-4-stilbazolium tosylate, crystalline polytetrafluoroethylene, or combinations thereof. In some embodiments, the density of the interlayer supports 150 may comprise about 5% to about 50% of the total area of the upper surface 126 of the substrate support layer 120 to provide a sufficient pore utilization increase to the composite 100, while at the same time minimizing a decrease in mechanical support of

the two-dimensional membrane layer 110. In some embodiments, the density of the interlayer supports 150 comprises 40 to 45 % of the total area of the upper surface 126 of the support substrate layer 120.

[0039] The interlayer supports 150 may be applied to the upper surface 126 of the support substrate layer 120 in any appropriate manner. For example, in some embodiments, the interlayer supports 150 may be carbon nanotubes that may be spray-coated in random orientations onto the upper surface 126 of the support substrate layer 120. The spray-coating may be controlled such that the density of the carbon nanotubes applied to the upper surface 126 may be fine-tuned. Other methods of disposing the interlayer supports 150 onto the upper surface 126 of the support substrate layer 120 may include, but are not limited to, electrostatic deposition, drop casting, spin-coating, sputtering, lithography, ion beam induced deposition, atomic layer deposition, or electron beam induced deposition. Additional methods include the application of electrospun fibers by an electric field, the acceleration of nanoparticles by a potential, or the use of an ion beam to irradiate material present on the upper surface 126 to form raised structures on the upper surface 126 of the support substrate layer 120.

[0040] FIG. 4 shows a cross-sectional view of the two-dimensional membrane layered structure 100 having flow passages 155 formed therein in accordance with a second embodiment. As shown in FIG. 4, before transferring the two-dimensional membrane layer 110 onto the support substrate layer 120, a plurality of grooves 150' are laterally formed onto the upper surface 126 of the support substrate layer 120 to form the flow channels 155. As such, fluid may flow through the pores 115b to the support substrate channels 125, thereby increasing the overall pore utilization of the two-dimensional membrane layer 110.

[0041] The plurality of grooves 150' may be formed into the upper surface 126 of the support substrate layer 125 by any appropriate means. For example, in some embodiments, the grooves 150' may be formed by etching the upper surface 126. In other embodiments, the grooves 150' may be formed by focused ion beam milling, lithography methods (e.g., optical, electron beam lithography, extreme UV lithography) on resists followed by suitable etching (e.g., reactive ion etching), block copolymer mask focused on columnar and aligned structures followed by suitable etching, laser ablation, nanoimprint, scanning probe

lithography, shadowmask deposition followed by suitable etching, or sparse aperture masking methods.

[0042] In some embodiments, the grooves 150' may be formed in a regular, structured pattern, such as a lattice-like pattern, or in a random pattern. In addition, the grooves 150' may be formed to have a depth and a width of about 1 nm to about 5 μm . In some embodiments, the depth and width may range from about 10 nm to about 1000 nm. In other embodiments, the depth and width of the grooves 150' may range from about 50 nm to about 250 nm. In certain embodiments, the density of the grooves 150' may comprise up to 50 to 75 % of the total area of the upper surface 126 of the support substrate layer 120.

[0043] Some embodiments, such as described above, allow for the formation of fluid flow passages in a two-dimensional membrane layered structure. In some embodiments, the flow passages may be formed by interlayer supports disposed on an upper surface of the support substrate layer. In other embodiments, the flow passages may be formed by grooves provided on the upper surface of the support substrate layer. In yet other embodiments, the flow passages may be formed both by interlayer supports disposed on the upper surface of the support substrate layer and grooves provided on the upper surface of the support substrate layer. The fluid flow passages may be formed between the two-dimensional support layer and the support substrate layer such that an increase in the amount of pores that allow fluid to flow to passages in the support substrate layer may be realized. Thus, the flow passages may result in an increase in overall pore utilization in the layered structure without resulting in a loss of mechanical support provided to the two-dimensional membrane.

[0044] The two-dimensional membrane layered structures described herein have broad application, including in water filtration, immune-isolation, (i.e., protecting substances from an immune reaction), timed drug release (e.g., sustained or delayed release), hemodialysis, and hemofiltration. Some embodiments described herein comprise a method of water filtration, water desalination, water purification, immune-isolation, timed drug release, hemodialysis, or hemofiltration, where the method comprises exposing a two-dimensional membrane layered structure to an environmental stimulus, and wherein the two-dimensional membrane layered structure comprises a two-dimensional membrane layer having a plurality of pores (e.g., a porous graphene-based material) and a support substrate layer having a plurality of substrate passages.

[0045] Some embodiments include methods of filtering water comprising passing water through a two-dimensional membrane layered structure. Some embodiments include desalinating or purifying water comprising passing water through a two-dimensional membrane layered structure. The water can be passed through the two-dimensional membrane layered structure by any known means, such as by diffusion or gravity filtration, or with applied pressure (e.g., applied with a pump or via osmotic pressure).

[0046] Some embodiments include methods of selectively separating or isolating substances in a biological environment, wherein the two-dimensional membrane layered structure separates or isolates biological substances based on characteristics of the substance, such as size. Such methods can be useful in the context of disease treatment, such as in the treatment of diabetes. In some embodiments, biological substances below a certain size threshold can migrate across the two-dimensional membrane layered structure. In some embodiments, even biological substances below the size threshold are excluded from migrating across the two-dimensional membrane layered structure due to functionalization of the plurality of pores, the plurality of substrate passages, the plurality of interlayer supports and/or the plurality of grooves.

[0047] In some embodiments, the plurality of pores, or at least a portion thereof, is functionalized. In some embodiments, the plurality of substrate passages, or at least a portion thereof, is functionalized, for instance by attaching or embedding a functional group. In some embodiments, the plurality of interlayer supports and/or the plurality of grooves, or at least a portion thereof, is functionalized, for instance by attaching or embedding a functional group. In some embodiments, the functionalization moieties are trapped between two layers, but are not restricted to a single position in the flow passages (i.e., they are mobile within the flow passages, but are inhibited from traversing the layers, e.g., based the size of the pores in the two-dimensional membrane layer). In some embodiments, functionalization comprises surface charges (e.g., sulfonates) attached to the pores, substrate passages, interlayer supports, and/or grooves. Without being bound by theory, it is believed that surface charges can impact which molecules and/or ions can traverse the two-dimensional membrane layered structure. In some embodiments, functionalization comprises specific binding sites attached to the pores, substrate passages, interlayer supports, and/or grooves. In some embodiments, functionalization comprises proteins or peptides attached to the pores, substrate passages, interlayer supports, and/or grooves. In some embodiments, functionalization comprises

antibodies and/or antigens (e.g., IgG-binding antigens) attached to the pores, substrate passages, interlayer supports, and/or grooves. In some embodiments, functionalization comprises adsorptive substances attached to the pores, substrate passages, interlayer supports, and/or grooves. In some embodiments, functionalization involves catalytic and/or regenerative substances or groups. In some embodiments, functionalization comprises a negatively or partially negatively charged group (e.g., oxygen) attached to the pores, substrate passages, interlayer supports, and/or grooves. In some embodiments, functionalization comprises a positively or partially positively charged group attached to the pores, substrate passages, interlayer supports, and/or grooves.

[0048] In some embodiments, functionalizing the pores, substrate passages, interlayer supports, and/or grooves functions to: restrict contaminants from traversing the two-dimensional membrane layered structure; act as a disposable filter, capture, or diagnostic tool; increase biocompatibility (e.g., when polyethylene glycol is used for functionalization); increase filtration efficiency; position the interlayer supports (e.g., interlayer supports can be positioned near the pores via affinity-based functionalization in the pores; additional spacers can be positioned in interlaminar areas); increase selectivity at or near the pores or in asymmetric two-dimensional membrane layered structure; and/or protect interlayer supports (e.g., from the external environment or from a particular vulnerability such as degradation).

[0049] Some embodiments have been described in detail with particular reference to preferred embodiments thereof, but it will be understood by those skilled in the art that variations and modifications can be effected within the spirit and scope of the claims.

WHAT IS CLAIMED IS:

1. A two-dimensional membrane layered structure comprising:
a support substrate layer having a plurality of substrate passages configured to allow fluid to flow therethrough;
a two-dimensional membrane layer disposed on an upper surface of the support substrate layer, the two-dimensional membrane layer having a plurality of pores configured to allow fluid to flow therethrough, the plurality of pores comprising a first portion of pores that overlap with the plurality of substrate passages and a second portion of pores that do not overlap with the plurality of substrate passages; and
a plurality of flow passages disposed between the support substrate layer and the two-dimensional membrane layer, the plurality of flow passages being configured to allow fluid to flow through the second portion of pores to the plurality of substrate passages.
2. The two-dimensional membrane layered structure of claim 1, further comprising a plurality of interlayer supports disposed on an upper surface of the support substrate layer, wherein the plurality of interlayer supports form the plurality of flow passages.
3. The two-dimensional membrane layered structure of claim 1, further comprising a plurality of grooves formed on an upper surface of the support substrate layer, wherein the plurality of grooves form the plurality of flow passages.
4. The two-dimensional membrane layered structure of claim 1, further comprising a plurality of interlayer supports disposed on an upper surface of the support substrate layer and a plurality of grooves formed on an upper surface of the support substrate layer, wherein the plurality of interlayer supports and the plurality of grooves form the plurality of flow passages.
5. A two-dimensional membrane layered structure comprising:
a support substrate layer having a plurality of substrate passages configured to allow fluid to flow therethrough;
a two-dimensional membrane layer disposed on an upper surface of the support substrate layer, the two-dimensional membrane layer having a plurality of pores configured to allow fluid to flow therethrough, and the plurality of pores comprising a first portion of

pores that overlap with the plurality of substrate passages and a second portion of pores that do not overlap with the plurality of substrate passages; and

a plurality of interlayer supports disposed on the upper surface of the support substrate layer,

wherein the plurality of interlayer supports is configured to form a plurality of flow passages that allow fluid to flow through the second portion of pores to the plurality of substrate passages.

6. The two-dimensional membrane layered structure of claim 5, wherein the plurality of interlayer supports comprises carbon nanotubes.

7. The two-dimensional membrane layered structure of claim 5, wherein the plurality of interlayer supports comprises electrospun fibers.

8. The two-dimensional membrane layered structure of claim 5, wherein the plurality of interlayer supports is disposed on an area of the upper surface of the support substrate layer ranging from about 5% to about 50% of the total area of the upper surface of the support substrate layer.

9. The two-dimensional membrane layered structure of claim 5, wherein the plurality of interlayer supports is functionalized.

10. The two-dimensional membrane layered structure of claim 5, wherein the plurality of interlayer supports is disposed on an area of the upper surface of the support substrate layer ranging from about 40% to about 45% of the total area of the upper surface of the support substrate layer.

11. A two-dimensional membrane layered structure comprising:
a support substrate layer having a plurality of substrate passages configured to allow fluid to flow therethrough;

a two-dimensional membrane layer disposed on an upper surface of the support substrate layer, the two-dimensional membrane layer having a plurality of pores configured to allow fluid to flow therethrough, and the plurality of pores comprising a first portion of pores that overlap with the plurality of substrate passages and a second portion of pores that do not overlap with the plurality of substrate passages; and

a plurality of grooves formed on the upper surface of the support substrate layer,
wherein the plurality of grooves is configured to form a plurality of flow channels that allow permeate to flow through the second portion of pores to the plurality of substrate channels.

12. The two-dimensional membrane layered structure of claim 11, wherein the plurality of grooves is formed on the upper surface of the support substrate layer in a lattice pattern.

13. The two-dimensional membrane layered structure of claim 11, wherein the plurality of grooves comprises a width of about 10 nanometers to about 1000 nanometers.

14. The two-dimensional membrane layered structure of claim 11, wherein the plurality of grooves is functionalized.

15. A method of increasing pore utilization in a two-dimensional membrane layered structure comprising:

providing a substrate support layer having a plurality of substrate passages configured to allow fluid to flow therethrough;

disposing a plurality of interlayer supports on an upper surface of the substrate support layer; and

disposing a two-dimensional membrane layer having a plurality of pores configured to allow fluid to flow therethrough on the upper surface of the substrate support layer having the plurality of interlayer supports, the plurality of pores comprising a first portion of pores that overlap with the plurality of substrate passages and a second portion of pores that do not overlap with the plurality of substrate passages,

wherein the plurality of interlayer supports form a plurality of flow passages, and

wherein the plurality of flow passages are configured to allow fluid to flow through the second portion of pores to the plurality of substrate passages.

16. The method of claim 15, wherein the plurality of interlayer supports comprises carbon nanotubes and wherein the carbon nanotubes are disposed on the upper surface of the support substrate layer via spray-coating.

17. A method for increasing pore utilization in a two-dimensional membrane layered structure comprising:

providing a substrate support layer having a plurality of substrate passages configured to allow fluid to flow therethrough;

forming a plurality of grooves on an upper surface of the substrate support layer; and

disposing a two-dimensional membrane layer having a plurality of pores configured to allow fluid to flow therethrough on the upper surface of the substrate support layer having the plurality of grooves, the plurality of pores comprising a first portion of pores that overlap with the plurality of substrate passages and a second portion of pores that do not overlap with the plurality of substrate passages,

wherein the plurality of grooves form a plurality of flow passages, and

wherein the plurality of flow passages are configured to allow fluid to flow through the second portion of pores to the plurality of substrate passages.

18. The method of claim 17, further comprising forming the plurality of grooves in a lattice pattern.

19. A two-dimensional membrane layered structure comprising:

a support substrate layer having a plurality of substrate passages configured to allow fluid to flow therethrough;

a two-dimensional membrane layer disposed on an upper surface of the support substrate layer, the two-dimensional membrane layer having a plurality of pores configured to allow fluid to flow therethrough, the plurality of pores comprising a first portion of pores that overlap with the plurality of substrate passages and a second portion of pores that do not overlap with the plurality of substrate passages; and

a flow passage means for allowing fluid to flow through the second portion of pores to the plurality of substrate passages to increase overall pore utilization of the two-dimensional membrane layer.

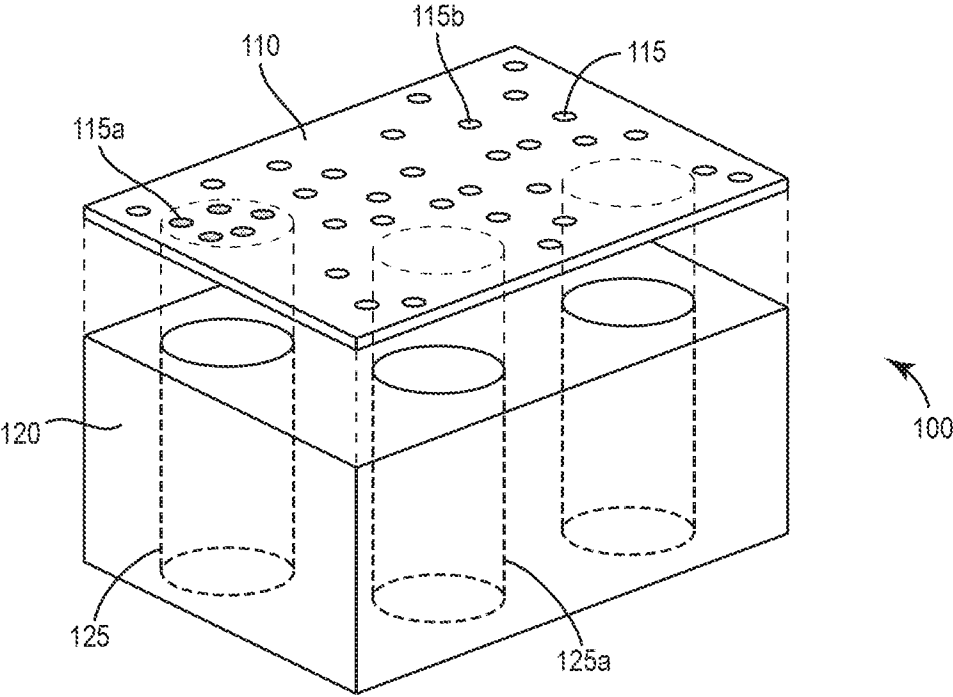


FIG. 1

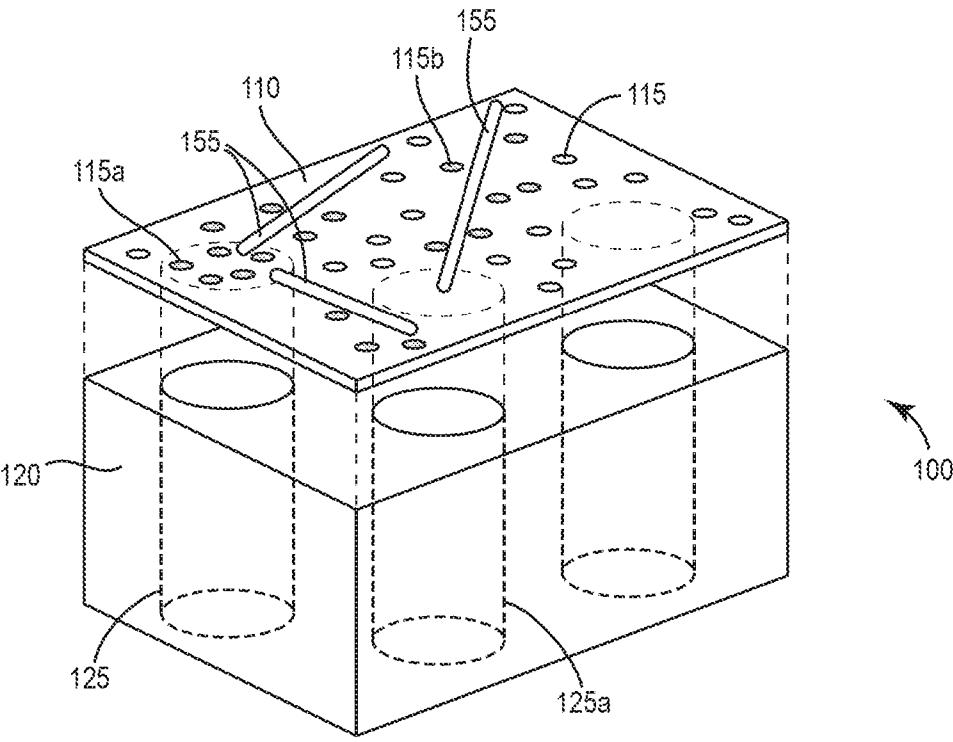


FIG. 2

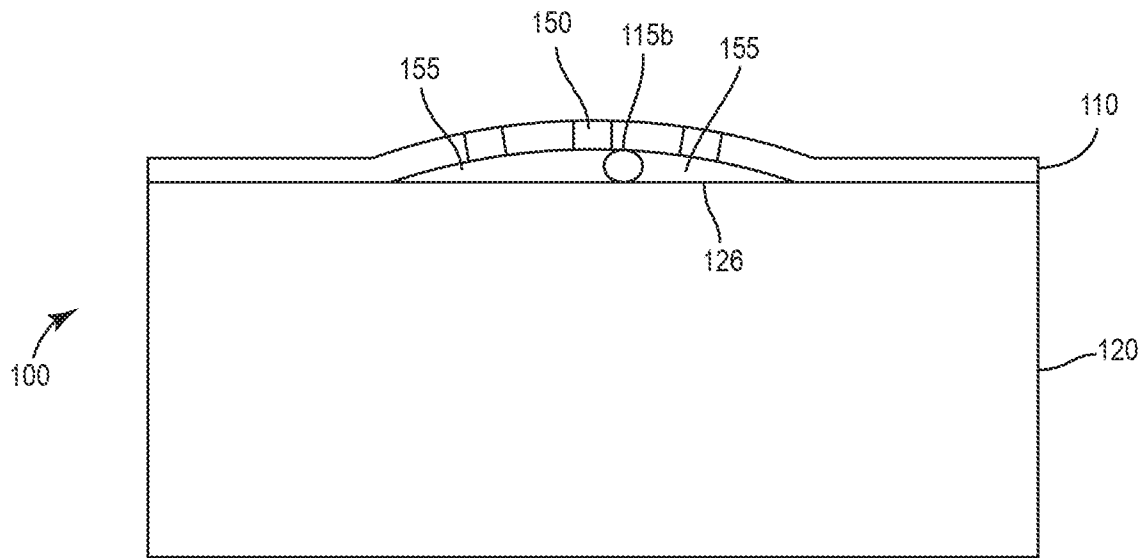


FIG. 3

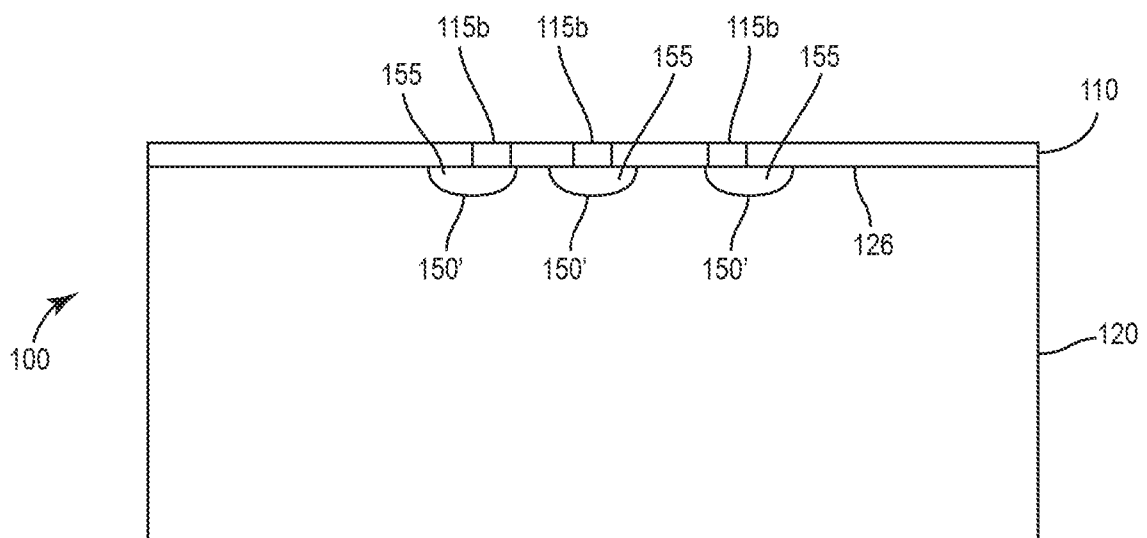


FIG. 4

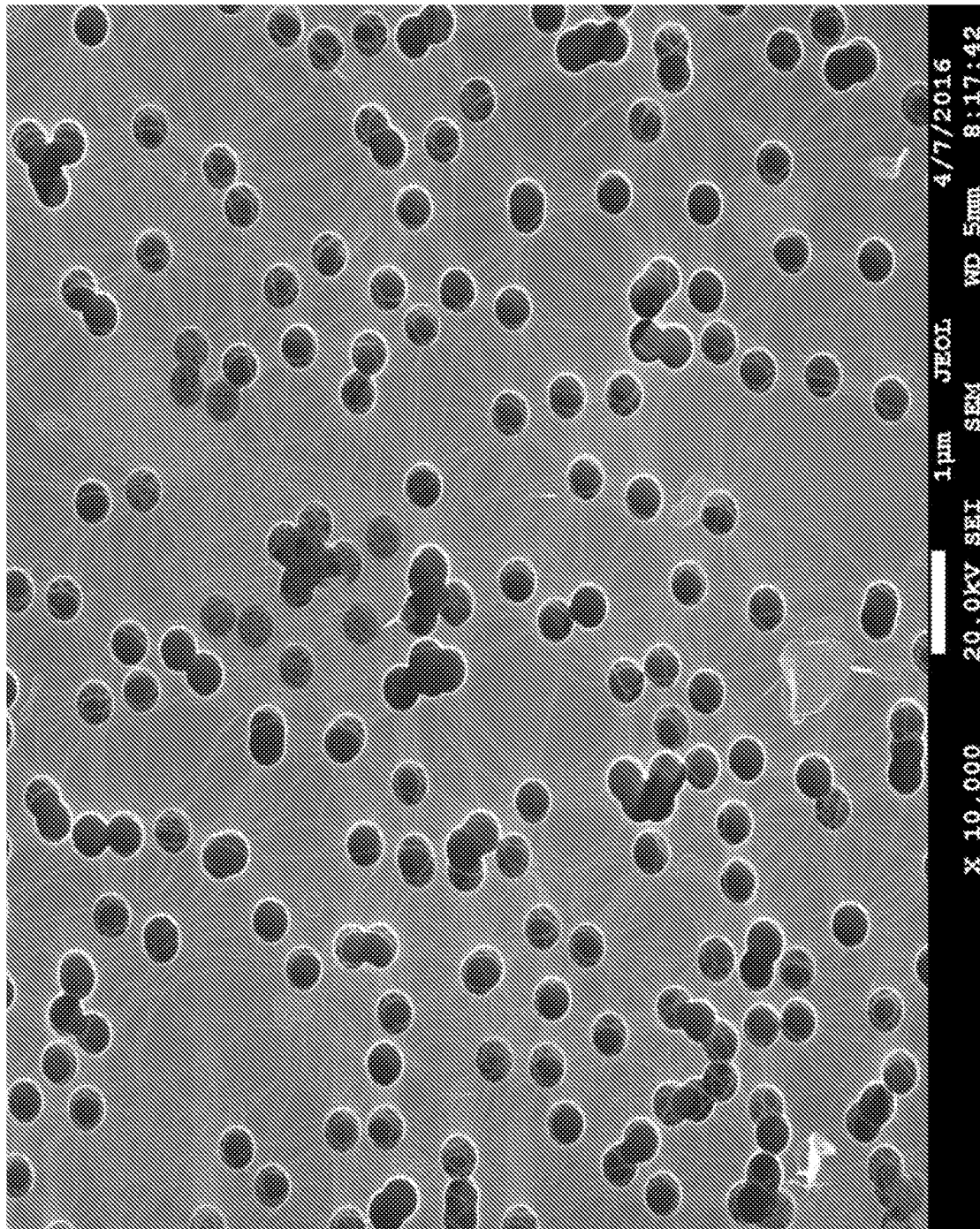


FIG. 5

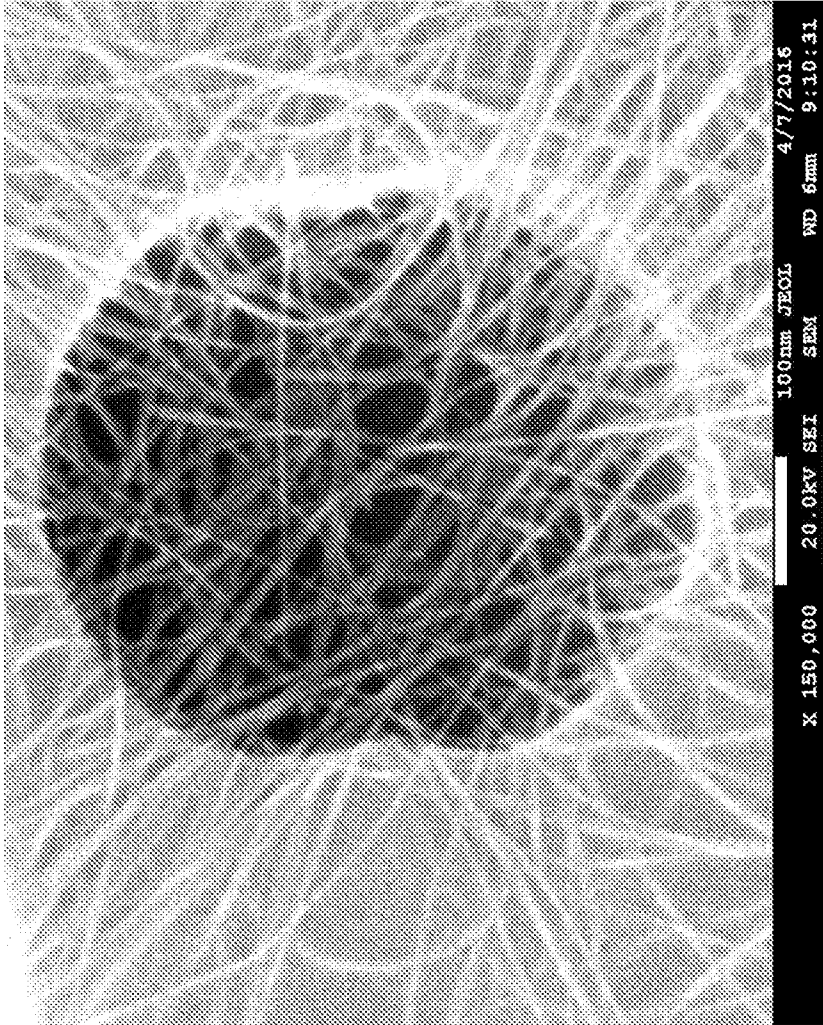


FIG. 6

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US2016/027629**A. CLASSIFICATION OF SUBJECT MATTER****B32B 3/20(2006.01)i, B32B 3/26(2006.01)i, B32B 3/30(2006.01)i**

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

B32B 3/20; B01D 65/00; C02F 1/44; B01D 63/08; B01D 61/00; B01D 71/02; B32B 3/26; B32B 3/30

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Korean utility models and applications for utility models

Japanese utility models and applications for utility models

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

eKOMPASS(KIPO internal) & Keywords:two-dimensional membrane, support, pore, passage, groove, interlayer, carbon nanotube

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2015-0321147 A1 (LOCKHEED MARTIN CORPORATION) 12 November 2015 See paragraphs [0012], [0015], [0026], [0081], [0083], [0094]; claims 1, 8, 17, 33, 35; and figures 13-15, 22, 23.	1-19
A	WO 2013-138698 A1 (MASSACHUSETTS INSTITUTE OF TECHNOLOGY et al.) 19 September 2013 See claims 1-4; and figure 1.	1-19
A	US 2015-0122727 A1 (MASSACHUSETTS INSTITUTE OF TECHNOLOGY et al.) 07 May 2015 See paragraphs [0094]-[0102]; and figures 7-10.	1-19
A	US 2013-0105417 A1 (LOCKHEED MARTIN CORPORATION) 02 May 2013 See paragraphs [0026]-[0029]; and figures 2-4.	1-19
A	US 2013-0256211 A1 (LOCKHEED MARTIN CORPORATION) 03 October 2013 See claim 1.	1-19



Further documents are listed in the continuation of Box C.



See patent family annex.

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

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