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(54) **AN APPARATUS FOR ANODIC OXIDATION OF VERY SMALL METAL GRIDS**

(57) The present invention belongs to the field of nanotechnology, more precisely to the field of nanostructures formed by manipulation of individual atoms, molecules, and to manufacturing processes for providing said nanostructures. The invention relates to an apparatus for anodic oxidation of small metal grids, which comprises:
- a housing comprising a bottom part and a top part arranged to receive a metal grid, wherein
o each part is provided with an opening for allowing contact with an electrolyte,
o one of the said parts is provided with a slot for receiving an electrical contact, said slot is arranged around the said opening,
o at least one, preferably at least one, preferably both

parts are provided with a slot for installation of a seal, preferably rubber seal, around the said opening,
o each part having assembly means to assemble both housing parts together,

- the electrical contact shaped as an elongated flat element with a circular head having a hole to receive the metal grid, said hole being slightly smaller than the metal grid to be anodized, wherein the elongated flat element is arranged to be connected to electricity, preferably using an alligator clip, and
- preferably the metal grid, typically a TEM grid usually having a diameter or length up to 5 mm.

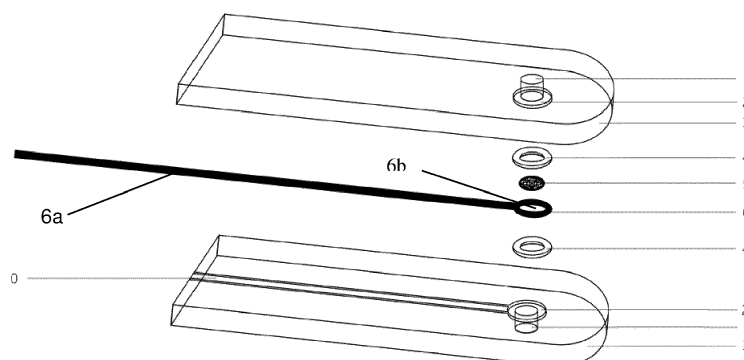


Figure 1

Description

Field of the invention

[0001] The present invention belongs to the field of nanotechnology, more precisely to the field of nanostructures formed by manipulation of individual atoms, molecules, and to manufacturing processes for providing said nanostructures. The invention relates to an apparatus for anodic oxidation of small metal grids.

Background of the invention and the technical problem

[0002] Various applications, such as synthesis of catalysts and active components in sensors, require immobilized nanostructured films. These can be the main catalysts or act as a high-surface-area support to immobilize active components in the form of different nanoparticles (e.g. metal nanoparticles) or organic molecules (e.g. enzymes).

[0003] Additionally, analysis of various samples with transmission electron microscope is done with TEM grids that can be purchased in different mesh size from 100 to 1000 and different materials such as copper, nickel, titanium, gold, molybdenum, etc., which can have a different coating, e.g. carbon or gold. The role of the coating is to provide an additional support on top of the grid to prevent samples from falling through the holes. The most common is a carbon coating, which provides support without interfering significantly with the flow of the electron beam. However, it is beneficial that the coating also has high surface area to which sample material can be attached.

[0004] There are several methods for synthesis of immobilized nanostructured films (nanotubes, nanoparticles, etc.) on a metal substrate, such as chemical vapour deposition (CVD), solvothermal synthesis, dip coating, but they are not appropriate for preparation of strongly attached nanostructured films on TEM grids since their size is too small. The most common dimension is 3.05 mm in diameter, the pitch of app. 25 μm to 250 μm . Additional disadvantages of the treatment of TEM grids with mentioned synthesis methods are their complexity and length of the process.

[0005] Another possible method for synthesis of nanostructures is anodic oxidation of metals which eliminates disadvantages of the above mentioned methods, but offers no appropriate anodization apparatus for the anodization of TEM grids due to their small size.

[0006] Hence, the technical problem solved by the present invention is provision of an apparatus for the preparation of an immobilized nanostructured film strongly attached to TEM grid with the anodic oxidation process in a simple and low-cost manner.

State of the art

[0007] The current state of the art comprises a wide variety of synthetic processes for the preparation of im-

mobilized, nanostructured materials, among which semiconducting materials such as TiO_2 , NiO and Cu_2O are very important and widespread. There are examples of the use of anodic oxidation process for the preparation of many different nanostructured films strongly attached to various metal shapes and forms (e.g. foil, mesh, wire, rod, etc.). However, there is no report on anodization of TEM grids or any metal elements with dimensions (e.g. diameter, length, width) smaller than 5 mm.

[0008] Liu et al. (2009; doi: 10.1021/jp903342s) describe growth of vertically oriented TiO_2 nanotube arrays on Ti meshes by electrochemical anodization. They started with 30-mesh or 50-mesh titanium samples which were folded along their diagonal and the two overlapped edges of each sample were cohered together using conductive silver paste. The double-layer Ti mesh was used as a working electrode. The Ti mesh was anodized at 60 V in electrolyte consisting of 0.25 wt.% ammonium fluoride and 2 vol.% milli-Q water in ethylene glycol.

[0009] Zeng et al. (2012; 10.1002/maco.201106481) synthesized the TiO_2 nanotube arrays on titanium mesh with electrochemical anodization in an electrolyte containing ammonium sulfate and ammonium fluoride. The 100-mesh titanium with the size of 30 mm $\times$ 55 mm and the thickness of 0.1 mm was polished and then anodized at 20 V for 20 minutes.

[0010] Sugiawati et al. (2019; doi: 10.3389/fphy.2019.00179) anodically oxidized titanium grids with dimensions of 1.3 cm $\times$ 1.3 cm, thickness of 0.57 mm and 0.23 mm wire diameter. The anodization electrolyte composition was glycerol with 1.3 wt.% NH_4F and 2 wt.% water. The titanium grid served as a working electrode and was pressed against an O-ring of the electrochemical cell leaving -0.63 cm^2 exposed to the electrolyte. Anodization was performed at constant voltage of 60 V and the anodization time was varied from 1 to 3 h.

[0011] Gerosa et al. (2016; doi: 10.1109/JPHOTOV.2016.2514702) prepared a nanostructured TiO_2 layer on the 60- μm -thick titanium grids with open area of 40% and the size of 1.4 cm $\times$ 1.4 cm. A TiO_2 layer was prepared by dipping the grid into the solution of commercial TiO_2 paste and isopropanol. The treated grid was left at room temperature for 10 min to allow the distribution of the TiO_2 solution on the whole surface. The last step was annealing at 525 $^\circ\text{C}$ for 30 min to crystallize the TiO_2 .

[0012] Gulati et al. (2015; 10.1021/acs.jpcc.5b03383) studied the anodization of curved titanium surfaces. The authors anodized titanium wires and reported on the role of electrolyte aging, water content, anodization voltage and time of anodization. The Ti wire with diameter 0.5 mm or 0.8 mm was immersed in the anodization electrolyte containing ethylene glycol, 0.3 wt. % ammonium fluoride and 1-3% deionized water.

[0013] None of these studies report on the special anodization setup. The anodic oxidations were performed in a two-electrode anodization cells in which the titanium anode was immersed in the electrolyte solution or

pressed against the anodization cell wall. There are also reports on sputtering different thin films on TEM grids. Lari, Steinhauer and Lazarov (2020; doi: 10.1007/s10853-020-04917-8) describe a method of sputtering amorphous Fe thin films on TEM carbon grids and preparation of Fe nanoparticles with treatment at elevated temperature. When the sample was heated to 600 °C, the thin film transforms into metallic Fe nanoparticles with a small presence of different Fe oxide nanoparticles. Further increase in the temperature to 700 °C resulted in the full oxidation of the NPs to magnetite.

[0014] Kim et al. (2005; 10.1017/S1431927605503106) describe the modification of the conventional method to prepare larger ultrathin carbon supporting film on TEM grids in order to maximize the thin film area and minimize the interference of the supporting film on the image. The carbon supporting film was prepared by immersing the TEM grids into a solution of 0.25 vol.% formvar in chloroform followed by slightly coating with carbon.

[0015] Patent applications CN101866804 (A) and US9406481 (B2) describe a method for application of carbon nanotubes on the TEM grids instead of traditional carbon coating. Similarly, CN107093543 (A) and CN109950117 (A) focus on the modification of TEM grids so that they become more appropriate for transmission electron microscopy investigations. For this purpose, graphene is deposited on TEM grids with different methods. Patents US8650739 and US9257258 describe a method for the fabrication of TEM grids.

[0016] Anodization of small metal meshes has not been described yet due to absence of suitable devices for anodization.

Description of the solution of the technical problem

[0017] The above-mentioned solutions are suitable for larger meshes and similar templates, diameters of which are larger than 1.3 cm. The invention addresses smaller meshes, having a diameter or length up to 5 mm, which pose a problem during anodization, as they tend to bend or otherwise mechanically deform. Furthermore, due to their small size it is a challenge to connect the mesh to electricity. The present invention is based on these considerations. The technical problem has been solved as defined in the independent claim, while preferred embodiments of the invention are defined in the dependent claims.

[0018] The invention relates to an anodization apparatus that enables the anodization of very small metal samples, e.g. TEM grids. Anodization of metals is a well-known process; however, anodization of very small metal grids has not yet been performed due to lack of suitable anodization apparatus for performing such anodization. TEM grids are the most common metal meshes and grids as they are widely used for transmission electron microscopy (TEM) analyses of various nanostructures. They can also be installed in very small devices such as mi-

croreactors or sensors. However, the invention is also suitable for other metal grids and meshes the largest dimension (e.g. diameter or length) smaller than 5 mm..

[0019] The essence of the present invention is the anodic oxidation apparatus comprising:

- a housing comprising a bottom part and a top part arranged to receive a metal grid, wherein
 - each part is provided with an opening for allowing contact with an electrolyte,
 - at least one, preferably both parts, are provided with a slot for receiving an electrical contact, said slot is arranged around the said opening,
 - at least one, preferably at least one, preferably both parts are provided with a slot for installation of a seal, preferably rubber seal, around the said opening,
 - each part having assembly means to assemble both housing parts together,
- the electrical contact shaped as an elongated flat element with a head, preferably circular head, said head having a hole to receive the metal grid, said hole being slightly smaller than the metal grid to be anodized, wherein the elongated flat element is arranged to be connected to electricity, preferably using an alligator clip, wherein the electrical contact is made from a thin metal foil, preferably cut with a laser into the desired shape, and
- preferably the metal grid with a diameter or length up to 5 mm, typically a TEM grid usually having a diameter between 1 and 5 mm.

[0020] The anodization apparatus, especially the housing, may be shaped in any suitable way depending on the operator performing anodization of very small metal grids. It is preferably shaped as an oval or a rectangle having one side with rounded corners and edges. The housing may be made from any non-conductive material; most preferably the housing is made of corrosion-resistant plastic. The assembly means may be any suitable, most preferably screws or magnets. They are preferably distributed to ensure that both parts of the housing are evenly pressed and thus ensure that the apparatus is sealed, so that only the grid is exposed to the anodization electrolyte. The number of assembly means depends on the shape of the housing, as well as the suitable distribution. For easier manipulation, the apparatus is preferably assembled together with magnets. In the simplified version, the apparatus can comprise only two parts; the bottom and top housing; which both include rubber seal and the electrical contact in each part of the housing.

[0021] The housing may also be formed to allow placement of two or more grids to be anodized. They can be placed side by side or one above the other. The electrical contact is provided for each grid in the placement side-by-side, while one electrical contact is sufficient for ver-

tical placement of grids if it is suitably adjusted.

[0022] The electrical contact and the seals can be permanently connected to the housing so that there are only two pieces, which are assembled together when a TEM grid is placed between them. The seals and electrical contact provide a flat surface where a TEM grid is placed so that it assures good electrical contact and no possibility for mesh bending. The electrical contact is an essential piece of the apparatus, as such small grids cannot be gripped directly using the alligator clip, as it could be dipped into the electrolyte, which hampers anodization, and/or too much surface would remain untreated (not anodized) and/or the grids could be bent, broken or in any other way mechanically damaged. The described design of the electrical contact, i.e. the elongated flat element with the suitably formed head allows the majority of grid's surface to be modified with a nanostructured catalytic film or nanostructured support for catalytic material- or other oxide film (e.g. compact). Said seals prevent the electrical contact from contact with the electrolyte into which the apparatus is dipped. The electrical contact has to be made from an inert material with good conductivity, usually it is made from any metal, preferably from steel, titanium or platinum. The seal protects the electrical contact to prevent oxidation. The hole in the head of the electrical contact may have any suitable shape to match the grid shape, usually it is circular, however, it may also be rectangular. The elongated flat element of the electrical contact is arranged to be connected to electricity, i.e. any suitable power source.

[0023] The apparatus can be used during anodic oxidation of small metal grids, said anodic oxidation can be performed with the setting of any suitable anodization parameters. The method for preparation of films, preferably nanostructured films, on the very small metal samples using the apparatus according to the invention preferably comprises the following steps:

- a) synthesis of an immobilized nanostructured film by anodic oxidation process which is done by: assembling the above-described anodization apparatus, placing it in an anodization cell with an anodization electrolyte and a cathode, applying electric potential between the electrodes and rinsing the grid and disassembling the apparatus after the anodization has finished,
- b) optionally annealing of the anodized film at elevated temperature that depends on the material that has been anodized (e.g. at 450 °C to convert the amorphous TiO_2 nanotubes into anatase crystal structure) to crystallize the nanostructured materials and improve the adhesion of the prepared film, and
- c) optionally post-treatment (e.g. additional heat treatment in specific atmosphere like NH_3 or H_2 , plasma treatment, hydrothermal treatment, etc.) that can be used to make the film catalytically active for various applications, increase the electric conductivity of the film or tune any other specific property

of the nanostructured film.

[0024] The electrolyte composition and temperature, anodization voltage and time, cathode material and other parameters can be chosen based on the metal grid material (e.g. titanium, copper, nickel, etc.) and the grid mesh size. The applied voltage can be in the range from 1 V to 100 V, preferably from 2 V to 50 V. Anodization time can be in the range from 30 seconds to 10 hours, preferably from 2 minutes to 1 hour. The temperature can be in the range from 0 °C to 180 °C, preferably from 20 °C to 30 °C. Cathode material can be any inert conductive material like platinum, stainless steel, titanium etc. Either one anodization or more can be performed.

[0025] The thickness of the film prepared by the anodization of metal grids depends on the metal that is anodized, its size (diameter of the wire that is forming the grid) and on the anodization parameters. The largest thickness is the same as the wire diameter, however, in this case the film mechanical properties are greatly deteriorated. The optimal thickness of the film depends on the intended use, which is obvious to the person skilled in the art.

[0026] The advantages of the method for the preparation of immobilized films with the described anodization apparatus include, among other things, the simplicity of the process, low price, short production time, great possibilities for changing the processing parameters and thus the properties of the resulting film, and the possibility of running the process continuously in an automated manner. The present invention successfully prevents bending of small meshes, TEM grids and similar templates or substrates for anodization, and at the same time provides the necessary space that allows connection of meshes to electricity. The apparatus according to the invention allows the meshes to be precisely positioned in the middle of the opening where the electrolyte enters.

[0027] The advantages of the apparatus according to the invention are:

- Simple anodization of very small meshes and grids,
- Highly homogenous nanostructured layer on the grids due to excellent electrical contact along the whole circumference of the grid,
- High-throughput anodization of grids if the housing can receive several grids, and
- Highly mechanically stable anodized grids, because bending of the grid is prevented by the housing.

[0028] The invention also relates to the nanostructured films prepared by the above-described method and the use of the so-prepared films in various applications, such as synthesis of catalysts for water and air purification, solar cells, electrolyzers, fuel cells, and active components in sensors, in which immobilized nanostructured films with large specific surface are required.

[0029] The importance of anodizing very small metal samples is not only due to their applicability in small cat-

alytic devices and sensors. The anodization of metal TEM grids also opens a new scientific field since the anodized metal grid enables the impregnation of high-surface-area support with catalytic nanoparticles and their simple characterization (e.g. electrochemical using floating electrochemical cell and morphological and structural using transmission electron microscopy).

[0030] The invention will be further disclosed and described based on exemplary embodiments and figures, which show:

Figure 1 Example of anodization apparatus that is used for the anodization of very small metal grids of different materials

Figure 2 Schematic presentation of the anodization cell that includes the special apparatus for anodization of metal grids shown in Figure 1

Figure 3 SEM micrograph of the top surface of anodized titanium TEM grid (A) which is made of immobilized TiO_2 nanotubes (B) strongly attached to the titanium metal grid substrate.

[0031] Figure 1 shows a possible embodiment of the anodization apparatus A, which comprises:

- A plastic housing 3 comprising a bottom part 3a and a top 3b part arranged to receive a metal grid 5, wherein
 - each part 3a,3b is provided with an opening 1 for allowing contact with an electrolyte,
 - One of the said parts 3a,3b is provided with a slot 0 for receiving an electrical contact 6, said slot is arranged around the said opening,
 - both parts 3a,3b are provided with a slot 2 for installation of a rubber seal 4, around the said opening,
 - each part having assembly means to assemble both housing parts together,
- the electrical contact 6 made from a thin metal foil shaped as an elongated flat element 6a with a circular head 6b having a hole 6b' to receive the metal grid 5, said hole 6b' being slightly smaller than the metal grid 5 to be anodized, wherein the elongated flat element 6a is arranged to be connected to electricity, preferably using an alligator clip, and
- the metal grid 5, typically a TEM grid usually having a diameter between 1 and 5 mm.

[0032] The scheme in Figure 2 shows the preferred embodiment of the apparatus and the anodic oxidation setup. The apparatus A is dipped into an anodization cell 10 filled with an electrolyte 11 and provided with a cathode 12. Power supply 13 is connected to the electrical contact 6 of the apparatus A, wherein the TEM grid 5 is placed in the circular hole of the electrical contact 6. The anodization cell is usually made of Teflon or other chem-

ically resistant plastics. The nanostructured film grows on the metal grid when a voltage of 1 V to 100 V is applied between the anode (metal grid) and the cathode. The time of the anodic oxidation can be between 30 seconds and 10 hours.

[0033] Figure 3 shows a SEM micrograph of the top surface of anodized titanium TEM grid (A). The result of anodic oxidation process is strongly attached TiO_2 nanotubular film (B). The 400-mesh TEM grid with diameter of 3.05 mm was anodized at 40 V for 20 minutes in anodization electrolyte with composition of 0.3 wt.% ammonium fluoride and 2 vol.% deionized water in ethylene glycol. The counter electrode was made of stainless steel.

[0034] Within the scope of the invention as described herein and defined in the claims, other embodiments of the method according to the invention that are clear to person skilled in the art may be possible, which does not limit the essence of the invention as described herein and defined in the claims.

Claims

1. An apparatus for anodic oxidation of small metal grids with the largest dimension, e.g. diameter or length, up to 5 mm, said apparatus comprising:
 - a housing comprising a bottom part and a top part arranged to receive a metal grid, wherein
 - each part is provided with an opening for allowing contact with an electrolyte,
 - one of the said parts is provided with a slot for receiving an electrical contact, said slot is arranged around the said opening,
 - at least one, preferably at least one, preferably both parts are provided with a slot for installation of a seal, preferably rubber seal, around the said opening,
 - each part having assembly means to assemble both housing parts together,
 - the electrical contact shaped as an elongated flat element made from a thin metal foil with a head having a hole to receive the metal grid, said hole being slightly smaller than the metal grid to be anodized, wherein the elongated flat element is arranged to be connected to electricity, preferably using an alligator clip.
2. The apparatus according to claim 1, wherein the metal grid is a TEM grid, said grid having a diameter between 1 and 5 mm, preferably 3.05 mm.
3. The apparatus according to claim 1 or claim 2, wherein the housing is shaped in any suitable way, preferably it is shaped as an oval or a rectangle hav-

ing one side with rounded corners and edges.

4. The apparatus according to any of the preceding claims, wherein the housing is made from any non-conductive material; most preferably of corrosion-resistant plastic. 5
5. The apparatus according to any of the preceding claims, wherein the assembly means may be any suitable, most preferably screws or magnets. 10
6. The apparatus according to any of the preceding claims, wherein the electrical contact and the seals are permanently connected to the housing so that there are only two housing parts, which are assembled together when the metal grid, preferably the TEM grid, is placed between them. 15
7. The apparatus according to any of the preceding claims, wherein the electrical contact has a circular or square head with a circular or a square hole, respectively. 20
8. The apparatus according to any of the preceding claims, wherein the electrical contact is made from an inert material with good conductivity, usually it is made from any metal, preferably from steel, titanium or platinum. 25
9. A method for preparation of immobilized films, preferably nanostructured films, on small metal grids using the apparatus according to any of the preceding claims, wherein the method comprises the following steps: 30
 - a) synthesis of an immobilized film by anodic oxidation process: 35
 - assembling the anodization apparatus,
 - placing it in an anodization cell with an anodization electrolyte and a cathode, 40
 - applying electric potential between the electrodes, and
 - rinsing the grid and disassembling the apparatus after the anodization has finished, 45
10. The method according to claim 9, wherein the method comprises further steps of:
 - b) optional annealing of the anodized film at elevated temperature that depends on the material that has been anodized to crystallize the prepared film and improve its adhesion, and 50
 - c) optional post-treatment that can be used to make the film catalytically active for various applications, increase the electric conductivity of the film or tune any other specific property of the film, preferably nanostructured film. 55

11. An anodized small metal grid with a largest dimension (e.g. diameter or length) up to 5 mm prepared with the apparatus according to any claim from 1 to 8 and/or the method according to claims 9 and/or 10.
12. The anodized small metal grid according to the preceding claim, **characterized in that** a highly homogeneous film, preferably nanostructured film is formed on its surface.
13. Use of the anodized small metal grid according to claim 11 or 12 as catalysts for water and air purification, solar cells, electrolyzers, fuel cells, and in sensors.

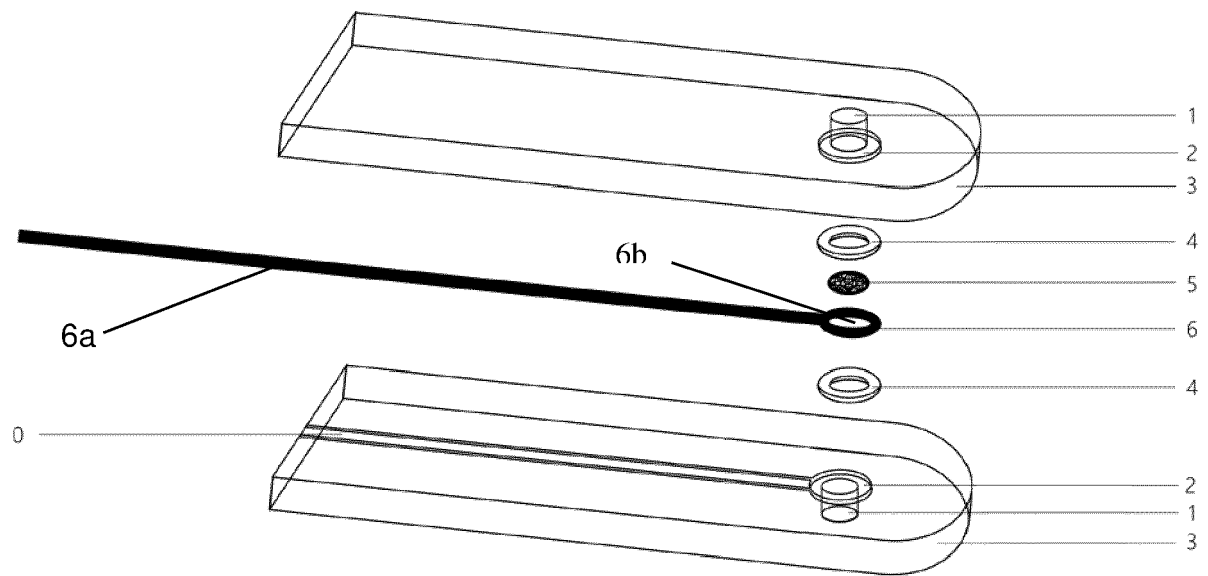


Figure 1

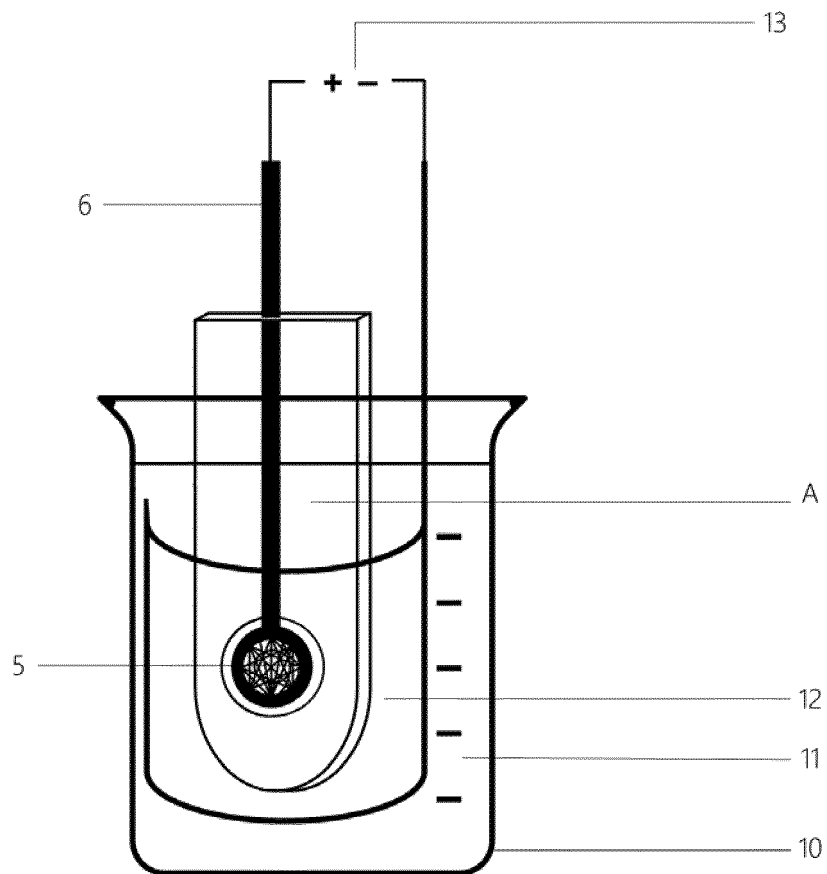


Figure 2

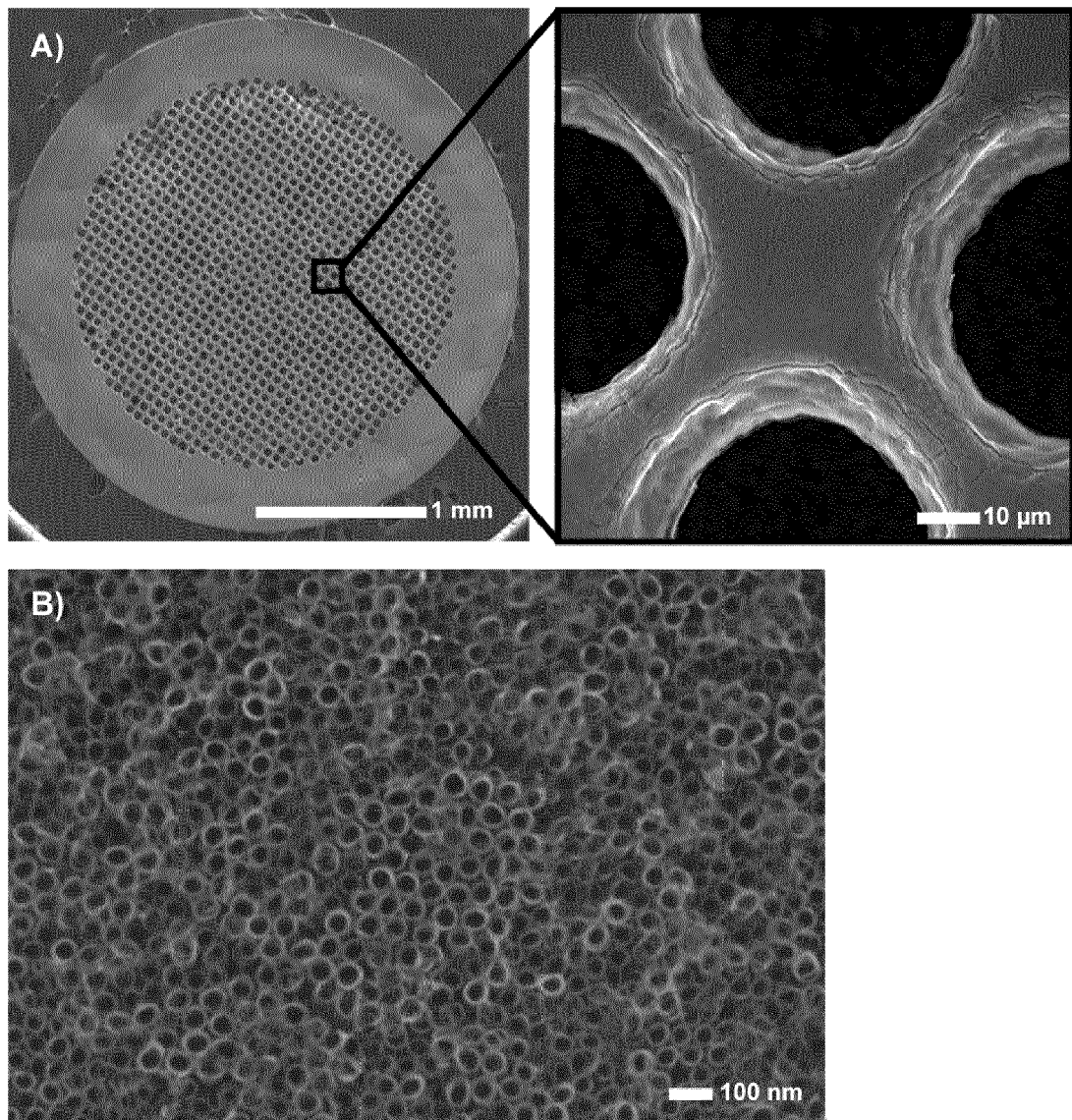


Figure 3



EUROPEAN SEARCH REPORT

 Application Number
 EP 21 16 3572

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The present search report has been drawn up for all claims			
Place of search The Hague		Date of completion of the search 30 August 2021	Examiner Lange, Ronny
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	

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EUROPEAN SEARCH REPORT

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Place of search The Hague		Date of completion of the search 30 August 2021	Examiner Lange, Ronny
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X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	

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**ANNEX TO THE EUROPEAN SEARCH REPORT
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EP 21 16 3572

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