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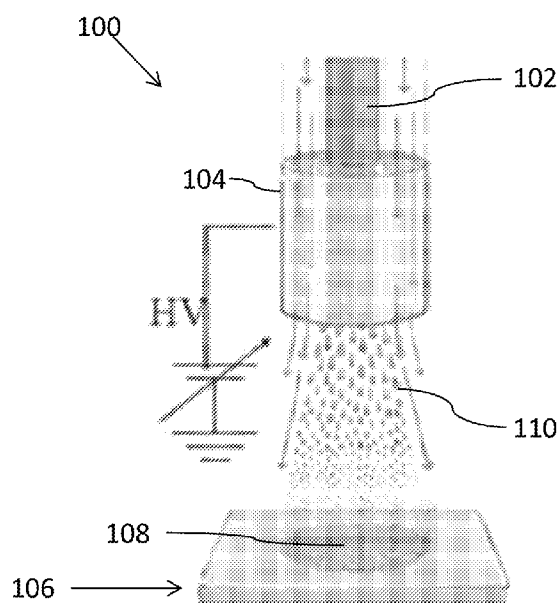


FIG. 2A

(57) Abstract: An electrospray apparatus (100) and a method for providing a coating layer over a substrate (108) are provided, which include: a capillary (104) having a spray area for ejecting a plurality of fluid samples; an air-flow system that is, at least in part, coaxially aligned with the spray area of the capillary; a collector (106) located, at least in part, spaced perpendicular to the capillary (104), the collector (106) comprising a substrate (108); and a voltage supply means that creates a potential difference between the spray area of the capillary (104) and the collector (106), the potential difference being sufficient to eject the plurality of fluid samples as a uniform stream of droplets (110), wherein circumferentially uniform air-flow from the air-flow system defines a size of each of the droplets (110).



FACILE ENCAPSULATION OF DYES VIA AIR-CONTROLLED ELECTROSPRAY**CROSS-REFERENCE TO RELATED APPLICATION**

[0001] The present application relates and claims priority to U.S. Provisional Application No. 62/466,739 filed March 3, 2017, the entirety of which is hereby incorporated by reference.

BACKGROUND OF THE INVENTION**1. Field of the Invention**

[0002] The invention relates generally to a system and method for nanoencapsulation and more particularly, nanoencapsulation of dyes via an air-controlled electrospray system.

2. Description of Related Art

[0003] In one aspect, mechanical coating techniques of substrates include various conventional techniques, such as roll coating, knife coating, etc. Disadvantageously, such mechanical coating techniques can often result in a surface coating having a non-uniform thickness differential that is greater than about 5 micrometers (μm) over the substrates. Further, the materials, such as solvents utilized during such processes can further contribute to the issues related to the uniformity due, at least in part, to issues, such as solubility, compatibility, stability and the like.

[0004] More recently, deposition techniques, such as electrospinning and electrospray techniques continue to be developed for use with coating of the substrates. By way of example, electrospray technique is a method of electrostatic atomization of a liquid or a solution to obtain charged micron-sized droplets, resulting in charged clusters and ions. For instance, the nozzle of the atomizer utilized in the electrospray process is typically made in the form of a metal capillary, which is biased by a high voltage. As understood by one skilled in the art, a solution of the material that is to be deposited onto the substrate is introduced into the metal capillary and the application of high voltage results in instability of the solution. The solution is then dispersed into small charged droplets that are about 0.3 to 20 microns in diameter. These droplets typically cover an upper surface of the substrate and coalesce to form a continuous coating over the substrate.

[0005] Further enhancements in electrospray processes, in particular, in the fabrication of novel materials with tailored nanostructures, continue to be pursued for enhanced performance and commercial advantage.

SUMMARY OF THE INVENTION

[0006] The present invention is directed to, inter alia, a method for providing a coating layer over a substrate, such as a nanofiber. In one embodiment, one or more fluid sample(s)

may be introduced into an electrospray apparatus which, for instance, may include a capillary that is charged to a high electric potential by a high power supply. By way of example, the fluid sample(s) may be, or may include, chemical components, such as a dye material, a polymer material, a solvent and the like. Each of these chemical components facilitates forming discrete droplets of a dye material that is encapsulated within a polymer material. According to an embodiment, each of these polymer-encapsulated dye droplets may be dispersed as capsules via the electrospray apparatus disclosed herein to provide a uniform coating layer over the nanofiber substrate. Further, in another embodiment, the electrospray apparatus includes an air-flow system that is, at least in part, coaxially aligned with the capillary so as to provide high-speed, circumferentially uniform air flow which can provide enhanced deformation of the droplets. Advantageously, such enhanced deformation of the droplets provides several benefits such as: (i) enhanced production rate (i.e., that is ten to hundred fold order of magnitude than that of conventional electrospray apparatus); (ii) better control of dispersion of nanoinclusions in each droplet; and (iii) better control of directing each droplet toward the collector so as to provide uniform sub-micron scale coating layers over the substrate.

[0007] In one aspect of the present application, an electrospray apparatus is provided. The electrospray apparatus includes: a capillary having a spray area for ejecting a plurality of fluid samples; an air-flow system that is, at least in part, coaxially aligned with the spray area of the capillary; a collector located, at least in part, spaced perpendicular to the capillary, the capillary including a substrate; and a voltage supply means that creates a potential difference between the spray area of the capillary and the collector, the potential difference being sufficient to eject the fluid as a uniform stream of droplets, wherein circumferentially uniform air-flow from the air-flow system defines a size of each of the droplets.

[0008] In another aspect of the present invention, a method for providing a coating layer over a substrate is provided. The method includes: providing a plurality of fluid samples disposed, at least in part, within an electrospray apparatus, the electrospray apparatus comprising: a capillary having a spray area for ejecting a plurality of fluid samples; an air-flow system that is, at least in part, coaxially aligned with the spray area of the capillary; a collector located, at least in part, spaced perpendicular to the capillary, the capillary including a substrate; and electrostatically atomizing the plurality of fluid samples to form a uniform stream of droplets, the plurality of fluid samples being electrostatically atomized in presence of circumferentially uniform air-flow from the air-flow system that defines a size of each of the droplets.

[0009] In yet another aspect of the present application, an electrospray apparatus is provided. The electrospray apparatus includes: a capillary having a spray area for ejecting a plurality of fluid samples; a collector located, at least in part, spaced perpendicular to the capillary, the capillary including a substrate; and a voltage supply means that creates a potential difference between the spray area of the capillary and the collector, the potential difference being sufficient to eject the fluid as a uniform stream of droplets.

[0010] In another aspect of the present invention, a method for providing a coating layer over a substrate is provided. The method includes: providing a plurality of fluid samples disposed, at least in part, within an electrospray apparatus, the electrospray apparatus comprising: a capillary having a spray area for ejecting a plurality of fluid samples; a collector located, at least in part, spaced perpendicular to the capillary, the capillary including a substrate; and electrostatically atomizing the plurality of fluid samples to form a uniform stream of droplets.

[0011] Additional features and advantages are realized through the techniques of the present invention. Other embodiments and aspects of the invention are described in detail herein and are considered a part of the claimed invention.

BRIEF DESCRIPTION OF THE DRAWINGS

[0012] One or more aspects of the present invention are particularly pointed out and distinctly claimed as examples in the claims at the conclusion of the specification. The foregoing and other objects, features, and advantages of the invention are apparent from the following description taken in conjunction with the accompanying drawings in which:

[0013] FIG. 1 is a schematic representation of a conventional electrospray system;

[0014] FIG. 2A is a schematic representation of an illustrative embodiment of an air-controlled electrospray system;

[0015] FIG. 2B is an alternative schematic representation of an illustrative embodiment of an air-controlled electrospray system;

[0016] FIG. 3 is a schematic representation of an illustrative embodiment of the air-controlled electrospray system for providing a coating layer over a nanofiber;

[0017] FIG. 4 depicts representative examples of scanning electron microscope (SEM) micrographs of each of one or more droplets having a polymer-encapsulated dye material that is dispersed via air-controlled electrospray system;

[0018] FIG. 5 depicts additional representative examples of scanning electron microscope (SEM) micrographs of a nanofiber coated with the dye encapsulated with polymer(s) via the air-controlled electrospray system, in accordance with one or more aspects of the present

invention.

[0019] FIG. 6 depicts a representative example of mechanochromic response of the nanofiber coated with the dye encapsulated with polymer(s) to compressive deformation; and

[0020] FIG. 7 depicts additional representative example of mechanochromic response of the nanofiber coated with the dye encapsulated with polymer(s) to extensional deformation.

DETAILED DESCRIPTION OF THE INVENTION

[0021] Aspects of the present invention and certain features, advantages, and details thereof, are explained more fully below with reference to the non-limiting examples illustrated in the accompanying drawings. Descriptions of well-known structures are omitted so as not to unnecessarily obscure the invention in detail. It should be understood, however, that the detailed description and the specific non-limiting examples, while indicating aspects of the invention, are given by way of illustration only, and are not by way of limitation. Various substitutions, modifications, additions, and/or arrangements, within the spirit and/or scope of the underlying inventive concepts will be apparent to those skilled in the art from this disclosure.

[0022] Referring now to FIG. 1, there is shown a conventional electrospray system. The depicted conventional electrospray system includes a capillary, which applies high voltage to a solution when the solution is injected into the capillary. The solution is atomized into fine charged droplets, as shown. However, also as shown in FIG. 1, the charged droplets produced by the conventional electrospray system are relatively large and are dispersed one at a time. Thus, conventional electrospray systems have a low throughput. Further, the dimensions, size, and geometries of the droplets dispersed are not controllable with the conventional electrospray system shown in FIG. 1. As a result, the enhanced size of each droplet produced by the conventional electrospray system inadvertently damages the substrate material on which it is electrosprayed. The damage is due, at least in part, to issues related to stress, such as compressive stress and/or tensile stress.

[0023] Turning now to FIGs. 2A-2B, there are shown a schematic representation of an illustrative embodiment of an air-controlled electrospray system 100. As shown in FIG. 2A, the air-controlled electrospray system 100 includes an electrospray apparatus 102 having a capillary 104 that is charged to a high electric (HV) potential, by high voltage (HV) power supply. As one skilled in the art will understand, a collector 106 that includes a substrate 108 is located, at least in part, spaced perpendicular to the capillary 104 during use of the air-controlled electrospray system 100. In some embodiments, the collector 106 that includes the substrate 108 may be located at an angle of about 10° to approximately 100° relative to the

capillary 104.

[0024] To begin the electrospraying process, one or more fluid samples are injected into the capillary 104 of the electrospray apparatus 102. The fluid sample(s) may be injected or otherwise inserted into the capillary 104 via a feeder (not shown), such as a syringe pump or a reservoir, either of which is attached, at least in part, to the electrospray apparatus 102. Further, the electrospray apparatus 102 may include an air-flow system that is, at least in part, coaxially aligned with the capillary 104, thereby defining an air-controlled electrospray apparatus 102. As understood by those of ordinary skill in the art, the air-flow system that is coaxially aligned with the capillary 104 advantageously provides high-speed, circumferentially uniform air flow which can provide enhanced deformation of the droplets 110 as disclosed further below.

[0025] In an alternative embodiment, shown in FIG. 2B, the polymer-encapsulated dye material is subjected to electrostatic atomization by exposing the chemical components to an intense electric field 112 (i.e., between the charged atomizer (e.g., capillary 104) and the collector 106) in the presence of high-speed, circumferentially uniform air-flow (that, for instance, is obtained from the air-flow system) to form the discrete polymer-encapsulate dye capsules. As understood, the charge transfer to the fluid and repulsive forces between the atomizer (e.g., capillary 104) and the fluid tear the droplets 110 from the atomizer (e.g., capillary 104) and send them toward the collector 106 in presence of uniform air-flow. As used herein, “electrostatic atomization” refers to conversion of the fluid material into very fine particles or droplets 110 by electrostatic forces

[0026] Referring now to FIG. 3, there is shown an illustrative embodiment of the air-controlled system 100 for providing a coating layer over a nanofiber. In the depicted embodiment, the fluid sample(s) injected into the capillary 104 of the electrospray apparatus 102 includes chemical components, such as a dye material (e.g., oil red), a polymer material (e.g., polystyrene and polyvinylidene fluoride), and a solvent. As understood, each of these chemical components is combined, at least in part, within the electrospray apparatus 102 to form the dye material that is encapsulated within the polymer material (referred to herein as “polymer-encapsulated dye material”).

[0027] In the embodiment shown in FIG. 3, the polymer-encapsulated dye material is electrostatically atomized to form discrete polymer-encapsulate dye droplets 110, which are then dispersed as capsules to provide a uniform coating layer over the substrate 108. In particular, in the depicted embodiment, the substrate 108 is a nanofiber, although, in alternative embodiments, the substrate 108 is a different webbing product or micro-fiber. To

form the polymer-encapsulated dye droplets 110 shown in FIG. 3, the fluid sample comprising the chemical components, such as the dye material (e.g., oil red), polymer material (e.g., polystyrene and polyvinylidene fluoride), and solvent is injected into the electrospray apparatus 102. The dye material is dissolved in the solvent, such as an organic solvent, for example. The chemical components are then electrosprayed with the air-controlled electrospray system 100. Atomization in the electrospraying process causes the solvent to evaporate. In this embodiment, the solvent is evaporated while the dye material is encapsulated within the polymer, which results in stretching (i.e., deforming) of each droplet 110 as it proceeds towards the collector 106. Such enhanced deformation of each droplet 110, advantageously, facilitates solidifying each droplet 110 into minute capsules that are deposited over the substrate 108 material. For example, in one embodiment, upon evaporation of the solvent, the polymer material migrates to the surface of the dye material, forming the polymer-encapsulated dye droplet 110, because the solvent is a poor solvent to the polymer.

[0028] The capsules produced by the air-controlled electrospray system 100, as shown in FIG. 3, are 70% smaller in diameter as compared to capsules produced by conventional electrospray systems. Accordingly, the capsules produced by the air-controlled electrospray system 100 can be sub-micron size. In an embodiment wherein the size of each of the droplets 110 may vary between microns to sub-micron sizes, the size depends upon various parameters, such as the flow rate of the chemical components being dispersed, applied voltage and fluid electric conductivity, and air-flow pressure. For example, the size of each of these droplets/capsules 110 is reduced by an order of magnitude of about micron to sub-micron size, owing to surface tension.

[0029] Further, as understood, the size of each droplet 110 is defined by the size of encapsulation of the dye material by the polymer material. Additionally, the high-speed, circumferentially uniform air-flow from the coaxially-aligned air-flow system (not shown) advantageously facilitates enhanced deformation of each droplet 110 by stretching and breaking the droplet 110 into minute capsules, thereby providing enhanced atomization of the polymer-encapsulated dye material. As multiple minute capsules may be formed virtually simultaneously, production of the capsules increases at least 10-fold with the air-controlled electrospray system 100. Further, as solvent evaporation occurs at a faster rate (as compared to that for conventional electrospray systems), the size and shape of the capsules is more uniform when the air-controlled electrospray system 100 is used.

[0030] Turning now to FIG. 4, there are shown representative examples of scanning electron microscope (SEM) micrographs of each of one or more droplets 110 having a

polymer-encapsulated dye material that are dispersed via air-controlled electrospray system 100. As understood, the SEM micrographs depicted in FIG. 4 reveal the size of each of these capsules that include polymer-encapsulated dye material. In one example, the dye material is encapsulated within polymers, such as, polystyrene and polyvinylidene fluoride. However, the exemplary polymers of FIG. 4 are presented by way of example only. Those skilled in the art will understand that other polymers, dye materials, and various other materials that are typically used with a conventional electrospray apparatus (in FIG. 1) could be employed for providing a coating layer over the substrate 108, such as nylon nanofibers or any conventional webbing products and/or micro-fibers.

[0031] In one embodiment by this approach, droplets 110 of the dye material encapsulated within a polyvinylidene fluoride polymer having an average diameter of about 3 μm are dispersed at a flow rate of about 0.009 mL/min, as depicted from the SEM micrographs depicted in FIG. 4. Note that, in this example, each of these droplets 110 exit from the capillary 104 (for instance, having a 16/12 gauge capillary tube with a diameter of about 18 cm) in the presence of an air-flow (i.e., from the co-axially aligned air-flow system) at a pressure of about 8 psi, at an applied voltage of about 12 kV.

[0032] In another example, by this approach, droplets 100 of the dye material encapsulated within a polystyrene polymer having an average diameter of about 0.96 nm are dispersed at a flow rate of about 0.03 mL/min, as depicted from the SEM micrographs depicted in FIG. 4. In this example, each of these droplets 100 exit from the capillary 104 (for instance, having a 20/10 gauge capillary tube with a diameter of about 20 cm) in the presence of an air-flow (i.e., from the co-axially aligned air-flow system) at a pressure of about 8 psi, at an applied voltage of about 18 kV. Note that, as further depicted in FIG. 4, the size distributions for each of the polystyrene-encapsulated dye capsule and polyvinylidene fluoride-encapsulated dye capsule are about 3 $\mu\text{m} \pm 1.3 \mu\text{m}$ and 0.96 nm $\pm 0.28 \text{ nm}$, respectively. As understood, the small droplet 110 size and their uniform size distribution resulted from the effective atomization of air-controlled electrospray where both controlled high-speed air stream and high electric field gradient enable synergistically breaking the jet into small uniform droplets 110. Further, in enhanced embodiment, each of the polymer-encapsulated dye material can be directly electrosprayed using the air-controlled electrospray apparatus 100 to provide a uniform coating layer over the substrates, such as, such as, nylon nanofibers or any conventional webbing products and/or micro-fibers, as depicted in FIG. 5.

[0033] Further, in an enhanced embodiment, the resulting polymer-encapsulated dye material that is incorporated in substrates 108, such as, nylon nanofibers (referred to herein as

“nanofiber composite material”) has been found to undergo compression. As depicted in FIG. 6, the rupture of polymer-encapsulated dye capsule increased with increase in the compressive force, revealing more of the dye material disposed therein. This effective mechanochromic response to compressive deformation may be used as an indicator of stress (for instance, such as, compressive stress) that each of the capsule that is exposed to in various safety equipment, such as, helmets, fall protection devices, fall restriction devices, fall arrest devices, fall restraint systems, work positioning devices, shock indicating devices, personal protection devices, harnesses, lifelines, anchorage systems, personal access systems, connecting devices, wear protection devices, rescue devices, webbing, rope and wire, descent devices, load arresting devices and the like.

[0034] As one skilled in the art will understand, “mechanochromism” refers to the change of color that occurs when chemicals, such as, polymer-encapsulated dye capsule/droplet are subjected to stress in their solid state by deformation, such as, mechanical grinding, crushing or milling, and the like.

[0035] Turning now to FIG. 7, there are shown additional representative example of mechanochromic response of the nanofiber coated with the dye encapsulated with polymer(s) to extensional deformation, in accordance with one or more aspects of the present invention. As understood, elongation of spirally wound polymer-encapsulated dye capsule incorporated, at least in part, within the substrate, such as, nanofiber leads to compression and rupture of each of these capsules. In one embodiment, the rupture of each of these capsules is dependent upon the binding and/or the strength, for instance, of the nanofiber, as evident from FIG. 7. As depicted, the capsules that include dye material encapsulated within the polystyrene polymer exhibited more sensitive rupture upon extension, possibly due to their large capsule sizes, while the sub-micro sized capsules that include the dye material encapsulated within the polyvinylidene fluoride polymer may withstand extensional deformation.

[0036] The terminology used herein is for the purpose of describing particular embodiments only and is not intended to be limiting of the invention. As used herein, the singular forms “a”, “an” and “the” are intended to include the plural forms as well, unless the context clearly indicates otherwise. It will be further understood that the terms “comprise” (and any form of comprise, such as “comprises” and “comprising”), “have” (and any form of have, such as, “has” and “having”), “include” (and any form of include, such as “includes” and “including”), and “contain” (any form of contain, such as “contains” and “containing”) are open-ended linking verbs. As a result, a method or device that “comprises”, “has”, “includes” or “contains” one or more steps or elements. Likewise, a step of method or an element of a

device that “comprises”, “has”, “includes” or “contains” one or more features possesses those one or more features, but is not limited to possessing only those one or more features. Furthermore, a device or structure that is configured in a certain way is configured in at least that way, but may also be configured in ways that are not listed.

[0037] The corresponding structures, materials, acts and equivalents of all means or step plus function elements in the claims below, if any, are intended to include any structure, material or act for performing the function in combination with other claimed elements as specifically claimed. The description of the present invention has been presented for purposes of illustration and description, but is not intended to be exhaustive or limited to the invention in the form disclosed. Many modifications and variations will be apparent to those of ordinary skill in the art without departing from the scope and spirit of the invention. The embodiment was chosen and described in order to best explain the principles of one or more aspects of the invention and the practical application, and to enable others of ordinary skill in the art to understand one or more aspects of the present invention for various embodiments with various modifications as are suited to the particular use contemplated.

CLAIMS

What is claimed is:

1. An electrospray system, the electrospray system comprising:
a capillary having a spray area for ejecting a plurality of fluid samples;
a collector located, at least in part, spaced perpendicular to the capillary, the collector comprising a substrate; and
a voltage supply means that creates a potential difference between the spray area of the capillary and the collector, the potential difference being sufficient to eject the plurality of fluid samples as a uniform stream of droplets.
2. The electrospray system of claim 1 further comprising an air-flow system that is, at least in part, coaxially aligned with the spray area of the capillary
3. The electrospray system of claim 2, wherein a circumferentially uniform air-flow from the air-flow system defines a size of each of the droplets.
4. The electrospray system of claim 1, wherein each of the plurality of fluid samples comprise at least one of a polymer and a solvent.
5. The electrospray system of claim 1, wherein the circumferentially uniform air-flow from the air-flow system facilitating enhanced deformation of each of droplet to form a polymer-encapsulated capsule.
6. The electrospray system of claim 1, wherein each of the plurality of fluid samples comprise a polymer, a solvent, and a dye material.
7. The electrospray system of claim 6, wherein the circumferentially uniform air-flow from the air-flow system facilitating enhanced deformation of each of droplet to form a capsule composed of the dye material encapsulated by the polymer.
8. A method for providing a coating layer over a substrate, the method comprising:
providing a plurality of fluid samples disposed, at least in part, within an electrospray apparatus, the electrospray apparatus comprising:

a capillary having a spray area for ejecting the plurality of fluid samples;

an air-flow system that is, at least in part, coaxially aligned with the spray area of the capillary;

a collector located, at least in part, spaced perpendicular to the capillary, the collector comprising the substrate;

electrostatically atomizing the plurality of fluid samples to form a uniform stream of droplets, the plurality of fluid samples being electrostatically atomized in presence of circumferentially uniform air-flow from the air-flow system to define a size of each of the droplets.

9. The method of claim 8, wherein the substrate is at least one of a nanofiber, a webbing product, and a micro-fiber.

10. The method of claim 8, wherein each of the plurality of fluid samples comprise at least one of a polymer and a solvent.

11. The method of claim 8, wherein each of the plurality of fluid samples comprise a polymer, a solvent, and a dye material.

12. The method of claim 11, further comprising the steps of:
dissolving the dye material in the solvent; and
evaporating the solvent such that the polymer forms an outer layer over the dye material.

13. The method of claim 8, the circumferentially uniform air-flow from the air-flow system facilitating enhanced deformation of each of droplet to form a polymer-encapsulated capsule.

14. A method for providing a coating layer over a substrate, the method comprising:

providing a plurality of fluid samples disposed, at least in part, within an electrospray apparatus, the electrospray apparatus comprising:

a capillary having a spray area for ejecting the plurality of fluid samples;

a collector located, at least in part, spaced perpendicular to the capillary, the collector comprising the substrate;

electrostatically atomizing the plurality of fluid samples to form a uniform stream of droplets.

15. The method of claim 9, further comprising an air-flow system that is, at least in part, coaxially aligned with the spray area of the capillary, wherein the plurality of fluid samples are electrostatically atomized in presence of circumferentially uniform air-flow from the air-flow system to define a size of each of the droplets.

16. The method of claim 14, wherein each of the plurality of fluid samples comprise a polymer, a solvent, and a dye material.

17. The method of claim 16, further comprising the steps of:
dissolving the dye material in the solvent; and
evaporating the solvent such that the polymer forms an outer layer over the dye material.

18. The method of claim 14, further comprising the step of injecting the plurality of fluid samples into the electrospray apparatus via a feeder attached thereto.

19. The method of claim 18, wherein the feeder comprises at least one of a syringe and a reservoir.

20. The method of claim 14, wherein the substrate is at least one of a nanofiber, a webbing product, and a micro-fiber.

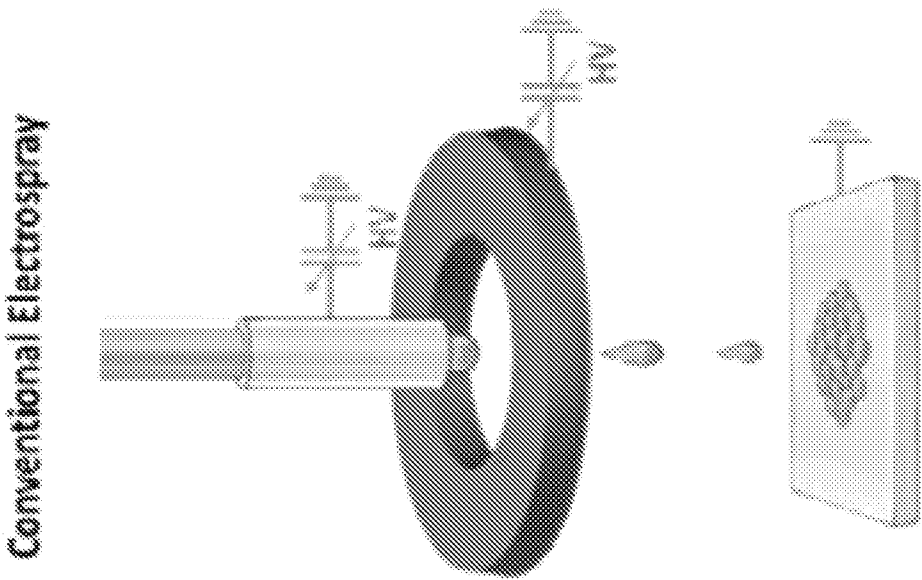


FIG. 1
(prior art)

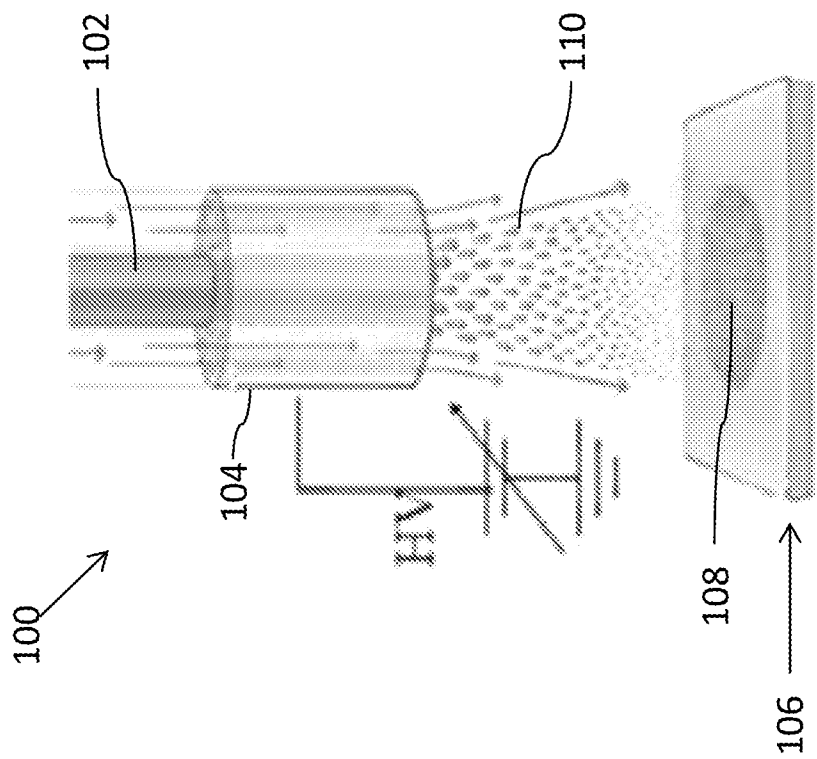


FIG. 2A

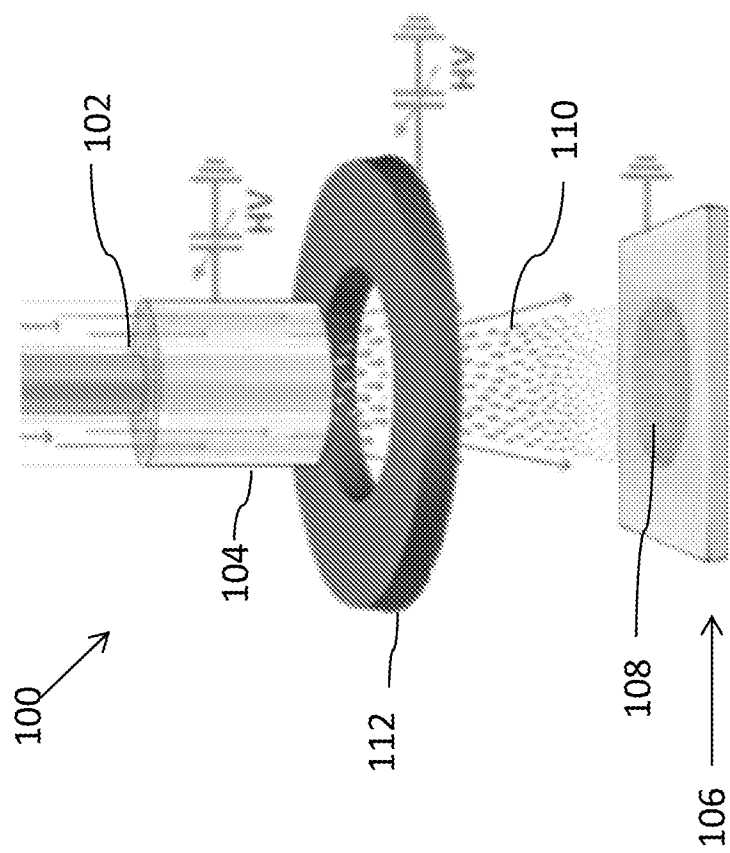


FIG. 2B

Air-controlled electrospray over Nylon nanofibers

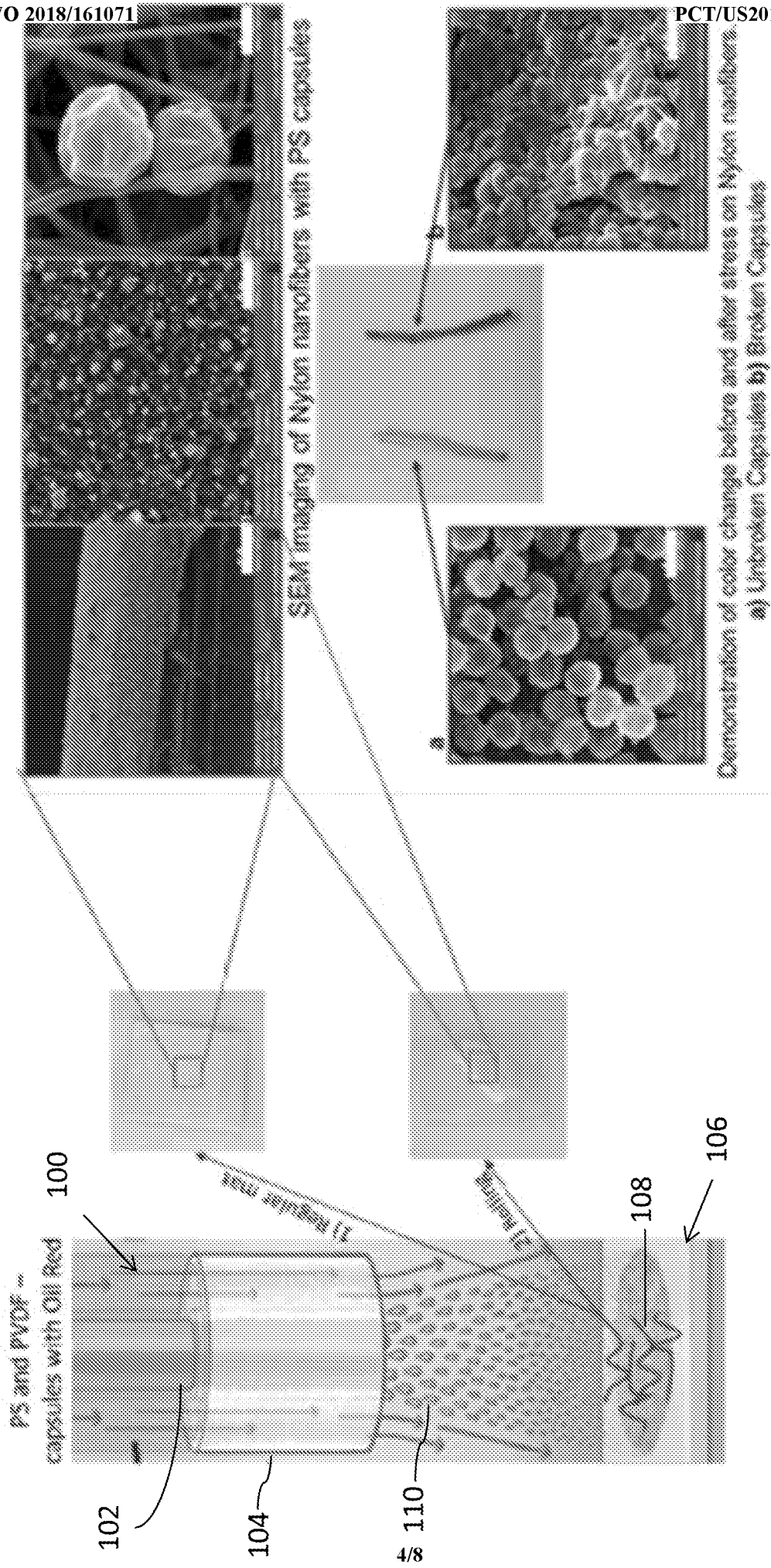
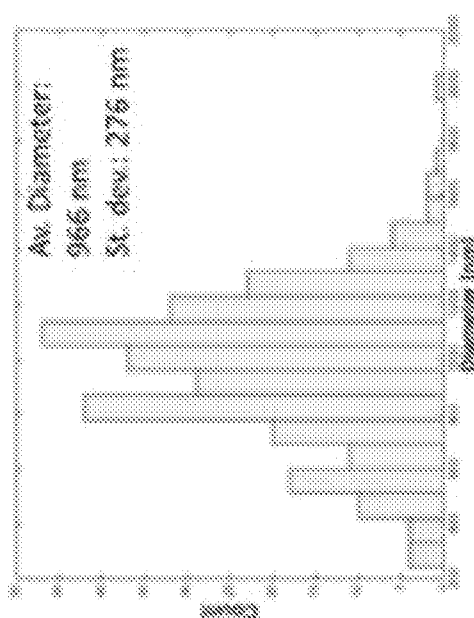


FIG. 3

Robust study to determine the best experimental conditions for PS/Oil Red and PVDF/Oil Red capsules

PVDF—12 kV, 0.009 ml/min, 1.8 cm, 16/12 image, 8 psi



Capacitance
Resistance
Impedance

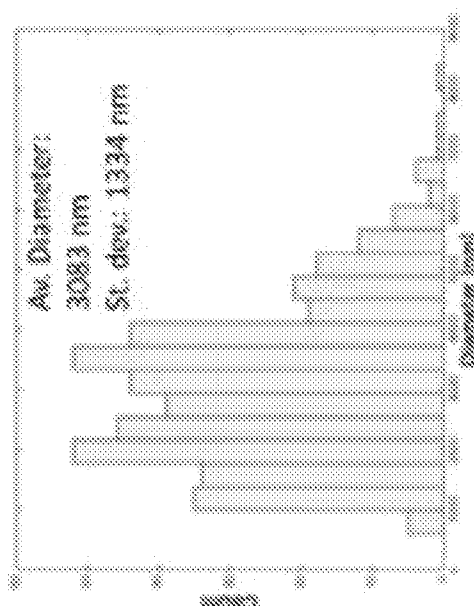


FIG. 4

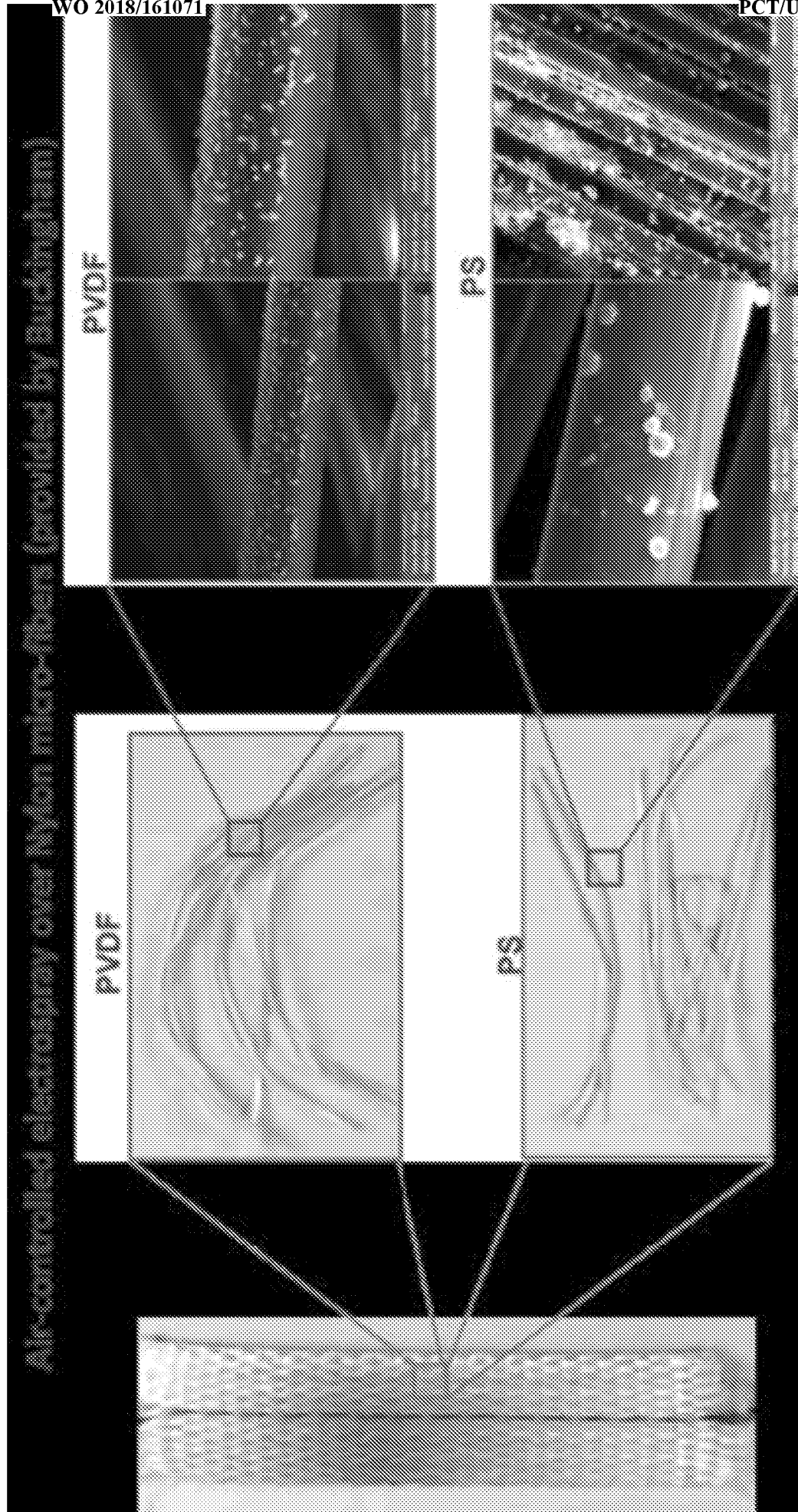


FIG. 5

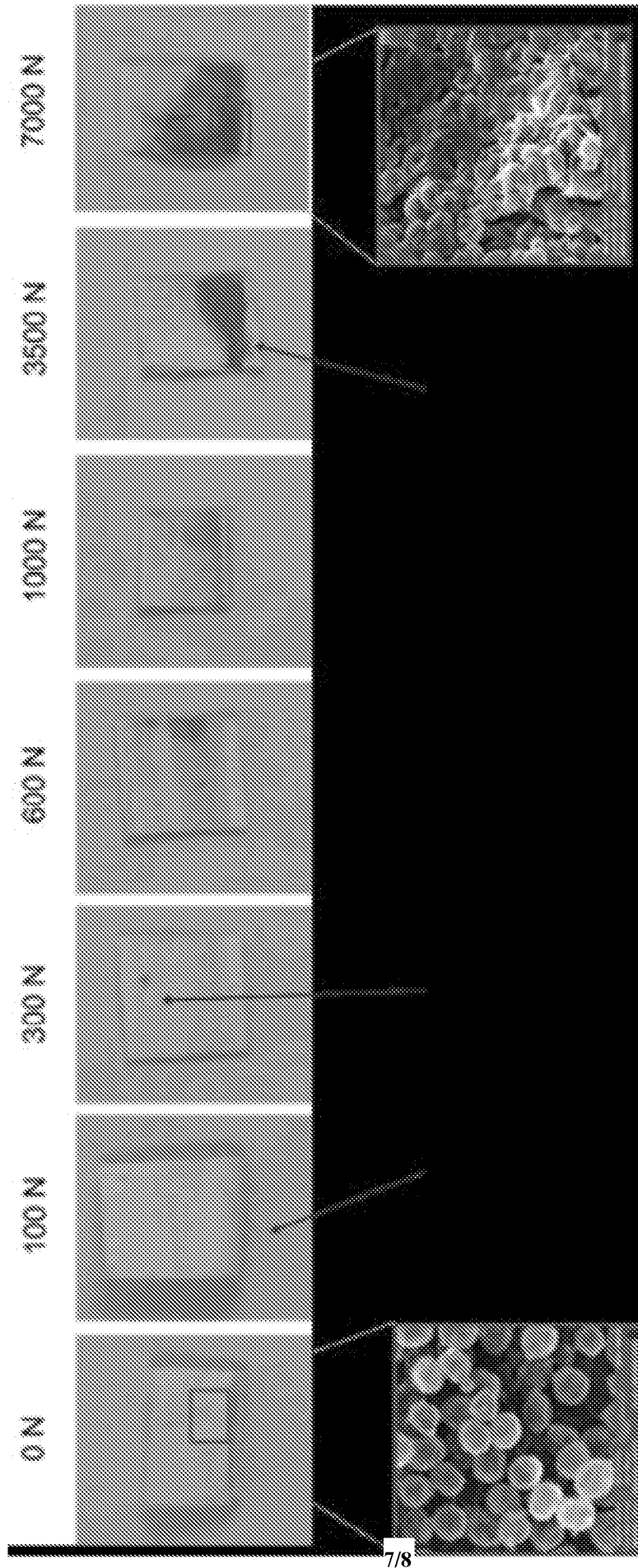


FIG. 6

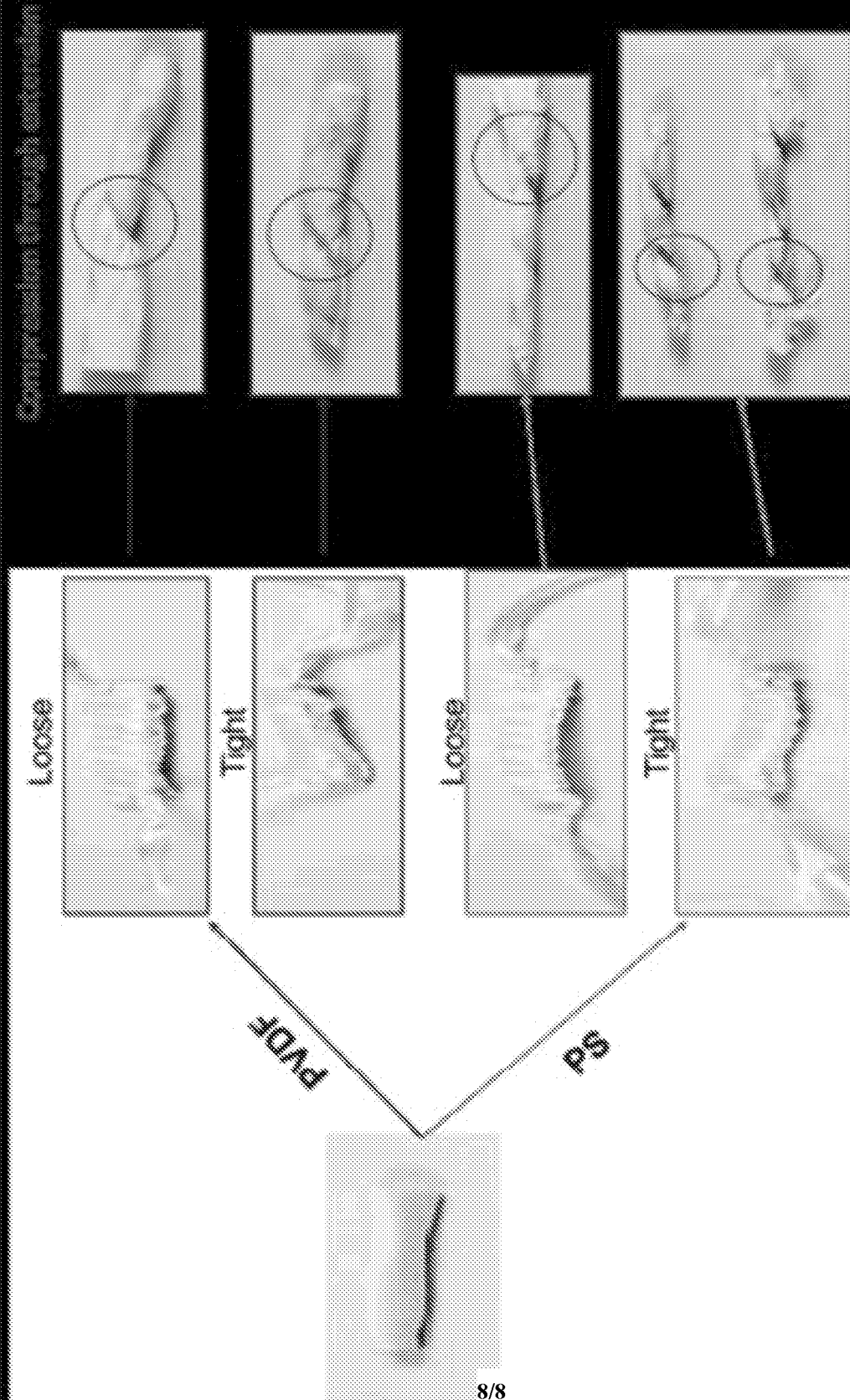


FIG. 7

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2018/020885A. CLASSIFICATION OF SUBJECT MATTER
INV. B05B5/03 B05B5/025
ADD.

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)
B05B

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

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

EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

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Y	the whole document	5-7, 11-13, 16,17
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Further documents are listed in the continuation of Box C.



See patent family annex.

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Date of the actual completion of the international search

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INTERNATIONAL SEARCH REPORT

International application No

PCT/US2018/020885

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

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International application No

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