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(54) SYSTEMS AND METHODS FOR ELECTRICAL ENERGY STORAGE

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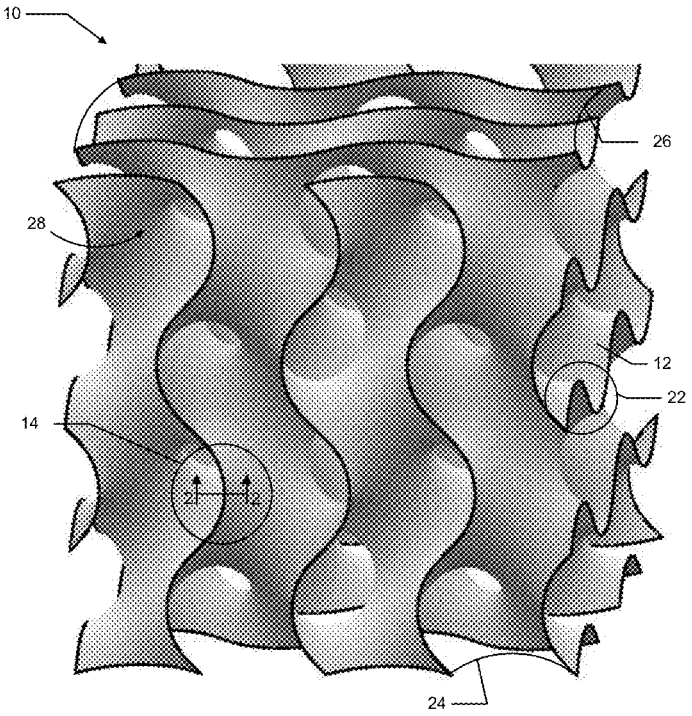
(57) **ABSTRACT**

The present disclosure relates to an electrical energy storage apparatus. The apparatus has an interpenetrating, three dimensional periodic structure formed from an ionically conductive solid electrolyte material having a plurality of interpenetrating, non-planar channels. The interpenetrating, non-planar channels are made up of a first plurality of channels filled with an anode material, a second plurality of channels adjacent the first plurality of channels and interpenetrating with the first plurality of channels, and filled with a cathode material, and a third plurality of channels adjacent to, and interpenetrating with, one of the first and second pluralities of channels, and filled with a material to form a separator. The first, second and third channels form a spatially dense, three dimensional structure. A first non-flat current collector layer is incorporated which is in communication with the first plurality of channels, and which forms a first electrode. A second non-flat current collector layer is incorporated which is in communication with the second non-planar channel, and which forms a second electrode.



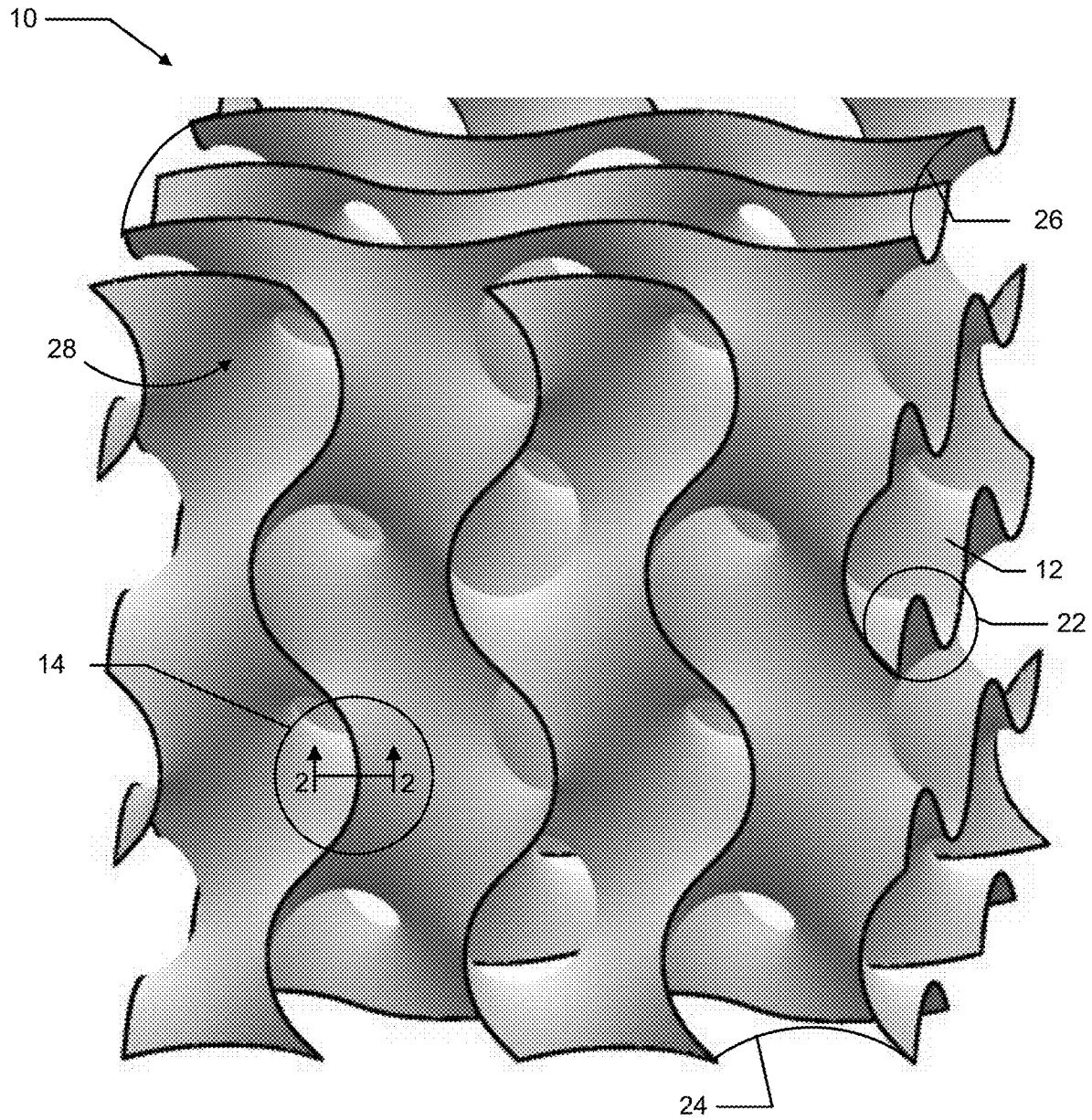


FIGURE 1

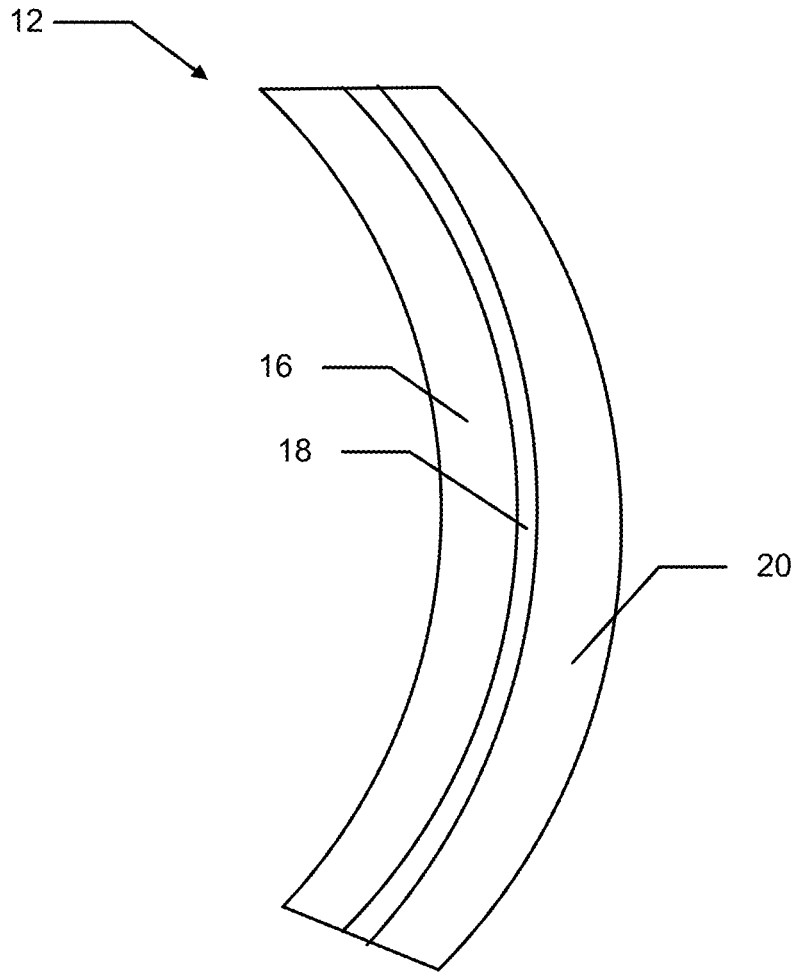


FIGURE 2

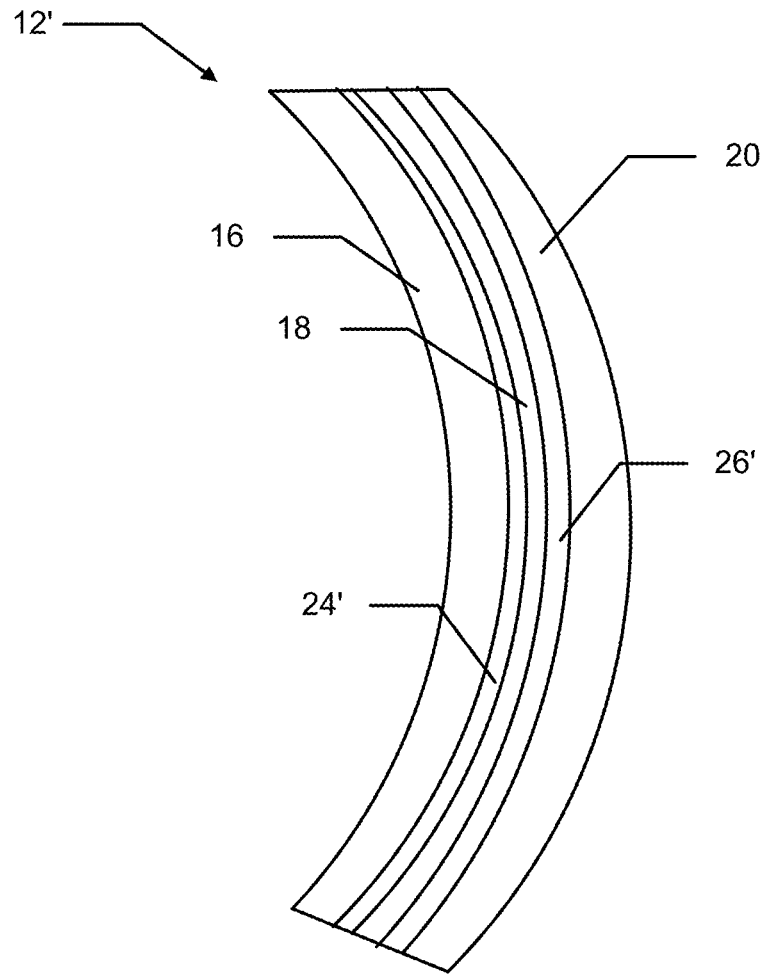
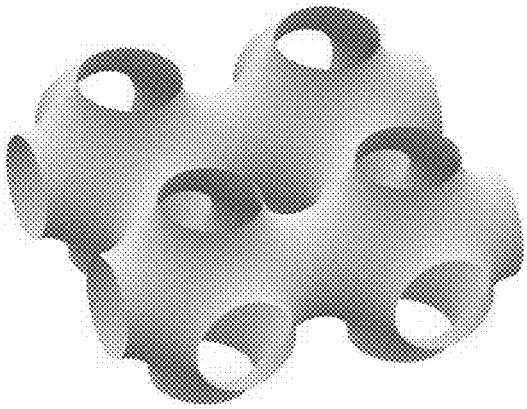


FIGURE 3

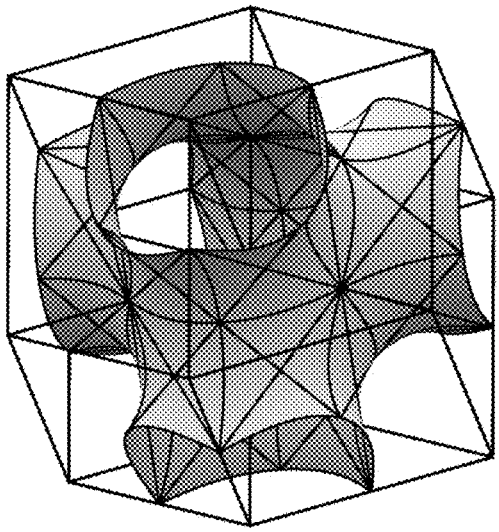
Schwartz P Surface

FIGURE 4



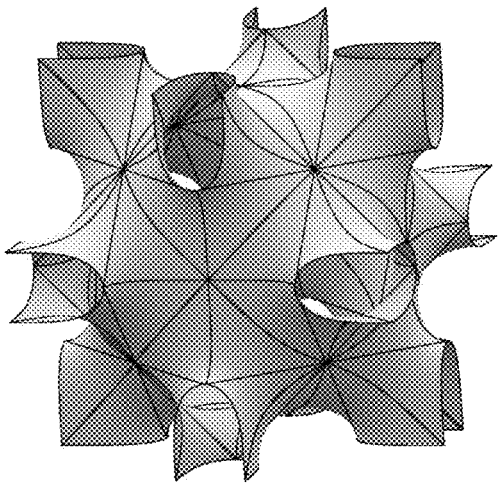
Schwartz D Surface

FIGURE 5



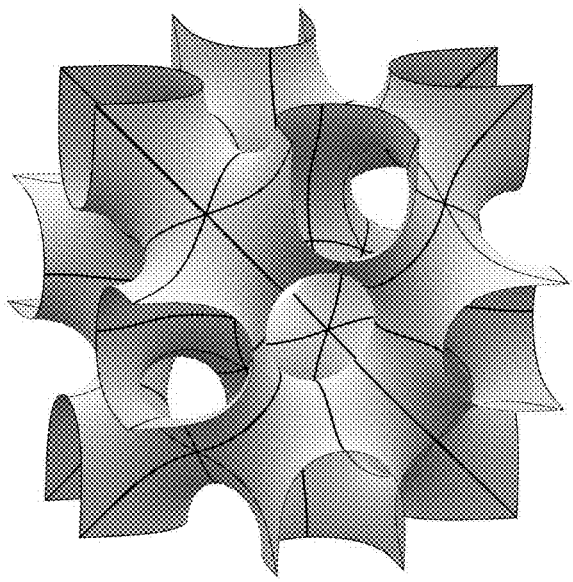
Neovius Surface

FIGURE 6



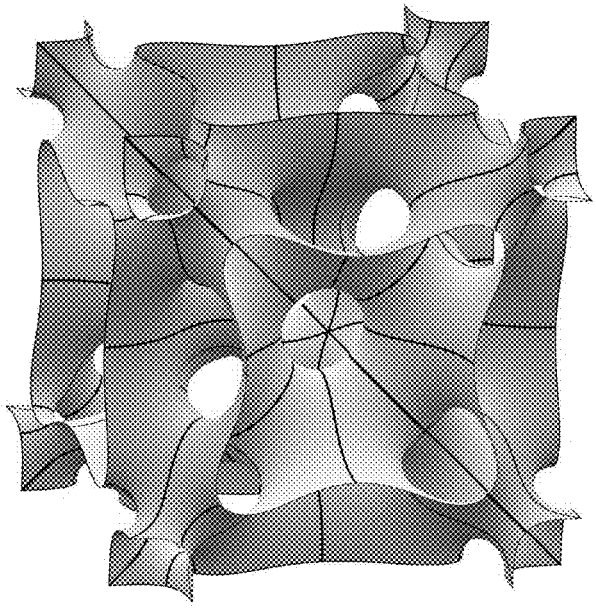
N14 Surface

FIGURE 7



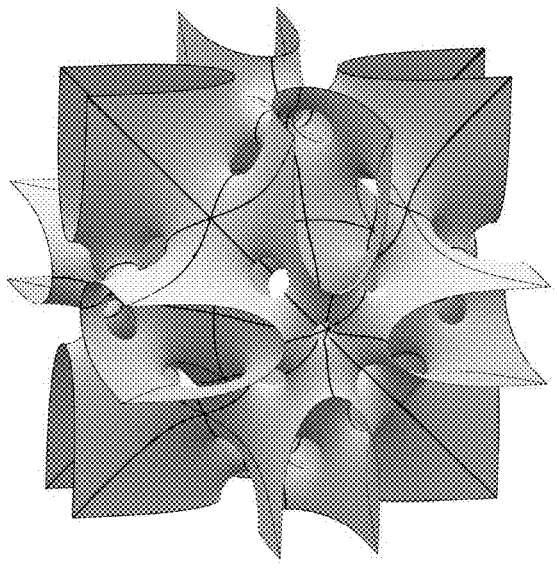
N26 Surface

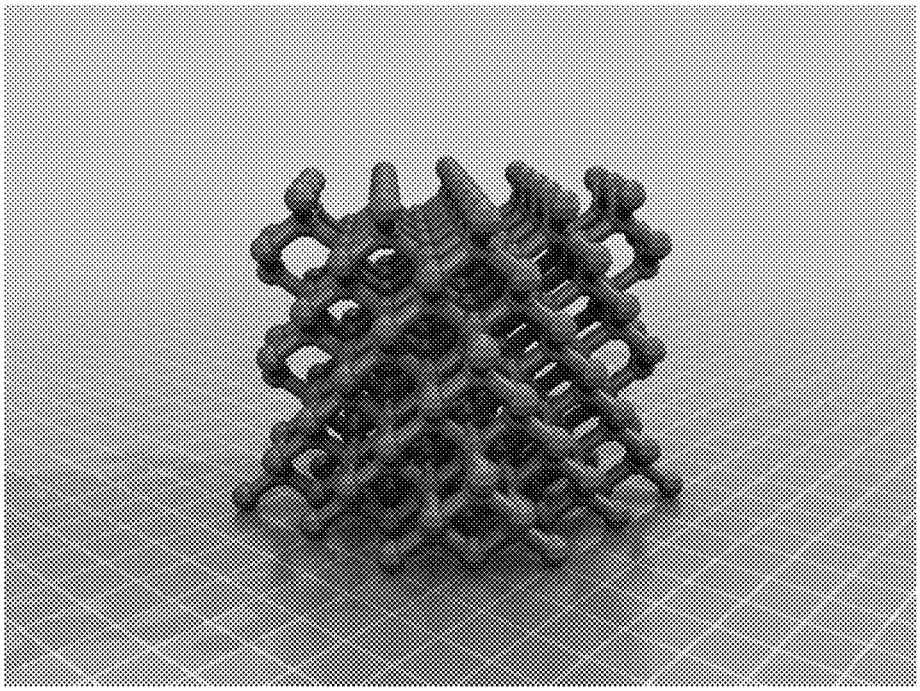
FIGURE 8



N38 Surface

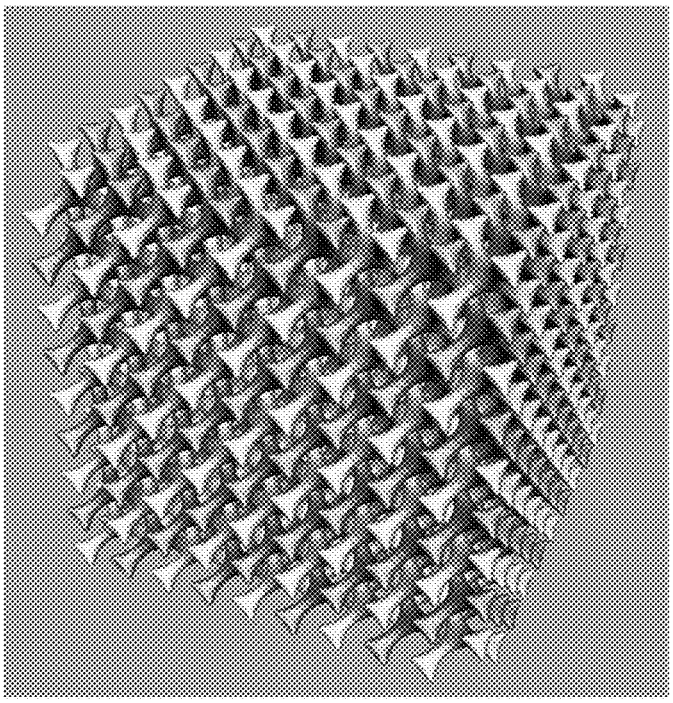
FIGURE 9





Diamond

FIGURE 10



Double Diamond

FIGURE 11

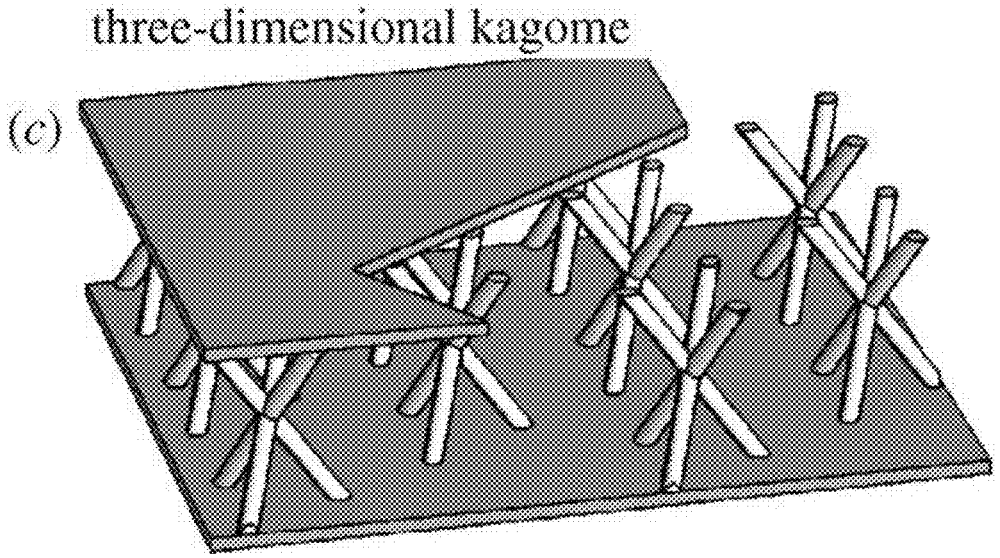


FIGURE 12

SYSTEMS AND METHODS FOR ELECTRICAL ENERGY STORAGE

CROSS-REFERENCE TO RELATED APPLICATIONS

[0001] This application is a divisional and claims priority of U.S. patent application Ser. No. 14/947,620 filed on Nov. 20, 2015. The entire disclosure of the above application is incorporated herein by reference.

FEDERALLY SPONSORED RESEARCH OR DEVELOPMENT

[0002] This invention was made with Government support under Contract No. DE-AC52-07NA27344 awarded by the United States Department of Energy. The Government has certain rights in the invention.

FIELD

[0003] The present disclosure relates to energy storage devices, and more particularly to highly penetrating, high surface area, three-dimensional structures for electrical energy storage.

BACKGROUND

[0004] The statements in this section merely provide background information related to the present disclosure and may not constitute prior art.

[0005] Over the past fifteen years or so, the proliferation of mobile electronics and electric vehicles has created an increasing demand for high-performance batteries that are lighter and store more energy on a single charge. Most improvements in battery technology have focused on achieving these objectives by developing new and better materials for the five main components of the battery: anode, cathode, conductive filler, electrolyte, and (if necessary) the separator. However, these previous efforts at improving battery technology have generally focused more on the materials used for the battery, but have largely ignored exploring the geometrical arrangement or internal "shape" or topology of a battery for the purpose of obtaining improvements in battery performance. There is also increasing interest in designing optimal internal micro or nanoscale structure for a macroscale object shape.

[0006] It will also be understood that conventional battery designs are generally planar. With a generally planar construction, the anode, separator and cathode of the battery are stacked on top of one another. These layers are then generally packaged in a planar form factor. Alternatively, these layers may be rolled up like a jelly roll and packaged into a cylindrical form factor.

[0007] Researchers have recently manufactured electrodes (i.e., anodes and cathodes) using geometries such as interdigitating combs or interdigitating posts. However, these efforts have not generally explored the possibility of increasing the performance of a battery by tailoring or controlling its physical geometry.

[0008] Some efforts have been made with regard to battery architecture, particularly involving designs using gyroid-like structures. U.S. Patent Publication No. 2014/0147747 discusses the construction of microbatteries using porous electrode architectures. U.S. Patent Publication No. 2014/0050988 discusses the use of gyroid structures (not any other minimal or triply periodic surfaces) specifically to form a charge collector. The charge collector is also known as a "current collector." This is the structure used to provide a path for electric current to or from the battery electrodes (anode and cathode).

SUMMARY

[0009] This section provides a general summary of the disclosure, and is not a comprehensive disclosure of its full scope or all of its features.

[0010] In one aspect the present disclosure relates to an electrical energy storage apparatus. The apparatus comprises an interpenetrating, three dimensional structure formed from an ionically conductive solid electrolyte material having a plurality of interpenetrating, non-planar channels. The interpenetrating, non-planar channels include a first plurality of channels filled with an anode material, a second plurality of channels adjacent the first plurality of channels and interpenetrating the first plurality of channels, and filled with a cathode material, and a third plurality of channels adjacent to, and interpenetrating with, one of the first and second pluralities of channels, and filled with a material to form a separator. The first, second and third channels form a spatially dense, three dimensional structure. A first non-flat current collector layer is incorporated which is in communication with the first plurality of channels, and which forms a first electrode. A second non-flat current collector layer is incorporated which is in communication with the second non-planar channel, and which forms a second electrode.

[0011] In another aspect the present disclosure relates to an electrical energy storage apparatus. The apparatus comprises an interpenetrating, three dimensional periodic structure formed from an ionically conductive solid electrolyte material having a plurality of interpenetrating, non-planar channels. The interpenetrating, non-planar channels include a first plurality of channels of an anode material, a second plurality of channels adjacent the first plurality of channels and interpenetrating with the first plurality of channels, and being of a cathode material, and a third plurality of channels adjacent to, and interpenetrating with, one of the first and second pluralities of channels, and being of a material to form a separator. A first current collector layer is incorporated which is in communication with the first plurality of channels, and which forms a first electrode. A second current collector layer is incorporated which is in communication with the second non-planar channel, and which forms a second electrode. The interpenetrating, three dimensional periodic structure further comprises one of: a gyroid; a double gyroid; a Schwartz surface; kelvin foam; an octet truss, a kagome lattice; a Neovius surface; an N14 Surface; an N26 Surface; an N38 Surface; a Diamond surface; and a Double Diamond surface.

[0012] In still another aspect the present disclosure relates to a method for forming an electrical energy storage apparatus configured as a three dimensional structure. The method comprises forming an interpenetrating, three dimensional periodic structure having a first plurality of non-planar channels and a second plurality of non-planar channels in proximity to the first plurality of non-planar channels. The first and second pluralities of non-planar channels are further formed to be interpenetrating. The method further includes filling each one of the first plurality of non-planar channels with an anode material to form an anode, filling each one of the second plurality of non-planar channels with a cathode material to form a cathode, and filling areas

adjacent to the first and second pluralities of non-planar channels with an electrolyte. The method further includes forming a first electrode to operate as a current collector, which is in electrical contact with portions of the anode material, and which forms a second electrode which is in electrical contact with portions of the cathode material.

[0013] Further areas of applicability will become apparent from the description provided herein. It should be understood that the description and specific examples are intended for purposes of illustration only and are not intended to limit the scope of the present disclosure.

BRIEF DESCRIPTION OF THE DRAWINGS

[0014] The drawings described herein are for illustration purposes only and are not intended to limit the scope of the present disclosure in any way.

[0015] FIG. **1** is a high level perspective view of a portion of a structure forming a 3D, periodic, energy storage device with interpenetrating wall portions, and where the 3D structure takes the form of a gyroid;

[0016] FIG. **2** is a cross sectional view of a portion of one of the material layers of the 3D structure of FIG. **1** taken in accordance with section line **2-2** in FIG. **1**;

[0017] FIG. **3** is a highly simplified view of a portion of a 3D solid electrolyte structure, made with a 3D printing process, having interpenetrating channels formed therein which are filled with anode and cathode materials;

[0018] FIG. **4** is a simplified illustration of a Schwartz P surface that may be used to construct a 3D energy storage apparatus in accordance with the present disclosure;

[0019] FIG. **5** is a simplified illustration of a Schwartz D surface that may be used to construct a 3D energy storage apparatus in accordance with the present disclosure;

[0020] FIG. **6** is a simplified illustration of a Neovius surface that may be used to construct a 3D energy storage apparatus in accordance with the present disclosure;

[0021] FIG. **7** is a simplified illustration of a N14 Surface that may be used to construct a 3D energy storage apparatus in accordance with the present disclosure;

[0022] FIG. **8** is a simplified illustration of a N26 Surface that may be used to construct a 3D energy storage apparatus in accordance with the present disclosure;

[0023] FIG. **9** is a simplified illustration of a N38 Surface that may be used to construct a 3D energy storage apparatus in accordance with the present disclosure;

[0024] FIG. **10** is a simplified illustration of a Diamond surface that may be used to construct a 3D energy storage apparatus in accordance with the present disclosure;

[0025] FIG. **11** is a simplified illustration of a Double Diamond surface that may be used to construct a 3D energy storage apparatus in accordance with the present disclosure; and

[0026] FIG. **12** is a simplified illustration of a Kagome lattice that may be used to construct a 3D energy storage apparatus in accordance with the present disclosure.

DETAILED DESCRIPTION

[0027] The following description is merely exemplary in nature and is not intended to limit the present disclosure, application, or uses. It should be understood that throughout the drawings, corresponding reference numerals indicate like or corresponding parts and features.

[0028] The various embodiments of the present disclosure generally relate to a class of microscale or nanoscale designs for three-dimensional, ("3D") structures. In one example the 3D structure is an electrical energy storage device, as will be described in detail herein. The 3D structure may be periodic or aperiodic. It may be ordered or disordered, but an important feature is that it is interpenetrating and 3D for all of the materials being used to form the structure. It could be graded density and feature sizes could change throughout the structure. As will become more apparent from the following discussion of a 3D energy storage device, as feature size decreases, the surface area increases and transport distances are reduced.

[0029] The 3D architectures disclosed herein are especially well suited for batteries where the anode, cathode, separator, electrolyte, and/or current collector are patterned into highly interpenetrating but discrete phases that have high surface areas and small transport distances while maximizing the amount of active material (i.e., anode or cathode) that can be packed into a given volume. The various embodiments disclosed herein have greater areal, volumetric, or gravimetric power density for a given energy density (or power density) compared to conventional battery designs based on planar layouts such as flat plates, jelly roll layouts, etc., or interdigitated geometries such as combs and posts. The power density may be limited by mass transport. The energy density is given by the nature and the packing density of the active material. As a result, for a given power load, the architectures disclosed herein may be used to manufacture batteries that last longer.

[0030] Referring to FIG. **1** there is shown a simplified representation of a 3D energy storage apparatus **10** (hereinafter simply "3D structure **10**") having interpenetrating layer portions that forms an electrical energy storage device. In this example the structure **10** takes the form of a gyroid, although it will be appreciated from the following discussion that a wide plurality of other 3D structures with interpenetrating walls or surface portions may be substituted for a gyroid. However, it will be appreciated that the anode, cathode, separator/electrolyte, and current collectors may or may not have all the same shape.

[0031] The 3D structure **10** of FIG. **1** includes 3D surface wall portions **12** which are formed relative to one another to be interpenetrating. By "interpenetrating" it is meant that one wall portion **12** cannot be disengaged from the other by any combination of translations or rotations. That is, in order to separate the two wall portions, which are not connected, one of the wall portions must be cut. Another example of an interpenetrating structure would be two links of a chain.

[0032] A small cross-sectional section **14** of just a portion of one of the surface wall portions **12** is shown in FIG. **2**. In FIG. **2**, surface wall portion **12** may be formed to include an anode material layer **16**, a separator material layer **18** and a cathode material layer **20**. The interpenetrating nature of the wall portions **12** can be noted, for example, at area **22**. It should be noted that it is impossible to go from anode material layer **16** to cathode material layer **20** without penetrating the separator material layer **18** (FIG. **2**).

[0033] With further reference to FIG. **1**, portions of all of the anode material layers **16** may be connected by an electrically conductive material layer or sheet **24**. Portions of all of the cathode material layers **20** may be connected by a separate electrically conductive material layer or sheet **26**. Material sheets **24** and **26** form current collectors, also

sometimes referred to as electrodes. The material sheets **24** and **26** have portions (not shown) where power connections can be made to some external device to allow stored electrical power from the 3D structure **10** to be used to power the external device. It will be understood that no portion of electrically conductive material sheet **24** contacts any of the cathode material layers **20**, and no portion of the material sheet **26** contacts any portion of the anode material layers **16**. These can be separated by a solid separator electrolyte or by a gap or void that is filled with liquid electrolyte. Liquid electrolytes are actually faster due to diffusion. In either event, when an electrolyte is used to fill areas **28**, this places the electrolyte in contact with all of the anode material layers **16** and all of the cathode material layers **20**, thus filling all of the voids within the 3D structure **10**.

[0034] The 3D surfaces used for patterning may be parametric. For a gyroid, for instance, boundaries of three-dimensional gyroid structures can be defined by the equations:

[0035] sin(2*pi*x/L)*cos(2*pi*y/L)+sin(2*pi*y/L)*cos(2*pi*z/L)+sin(2*pi*z/L)*cos(2*pi*x/L)=+t/2 and sin(2*pi*x/L)*cos(2*pi*y/L)+sin(2*pi*y/L)*cos(2*pi*z/L)+sin(2*pi*z/L)*cos(2*pi*x/L)=−t/2 [6], so that the thickness of the gyroid is the parameter "t" and its period (i.e., the length of a unit cell) is "L".

[0036] Controlling the thickness of the surface wall portions **12** tunes ion transport properties so that active material is depleted from the anode material layer **16** evenly. Consequently, for different active materials, the thickness of the surface(s) used in the design may change. In general, thinner is better. Ideally, the active materials should have a nanoscale thickness.

[0037] It is also expected that manufacturability constraints are likely to also place constraints on the thickness of the surface, as well as its unit cell length.

[0038] A 3D electrical energy storage structure such as 3D structure **10** in FIG. **1** may be manufactured using present day 3D printing or 3D fabrication processes. If manufactured using a well known 3D printing process, then the 3D structure **10** will be manufactured as a series of discrete layers successively formed one on top of another. In this fashion the channels necessary to form the anode layer, the cathode layer, and any other material layers (e.g., separator layer) would be formed substantially simultaneously as each layer is printed when the different types of material are deposited by different print heads of a 3D printing system.

[0039] FIG. **3** shows one high level example of how the charge collectors may be formed with an interpenetrating construction. In this example the wall portion **12'** has charge collectors **24'** and **26'** disposed in interpenetrating fashion on opposite surfaces of the separator layer **18**. Charge collector **24'** is disposed in interpenetrating fashion with anode material layer **16** and charge collector **26'** is disposed in interpenetrating fashion with cathode material layer **20**. Such a construction minimizes electrical transport distances to the charge collector layers **24'** and **26'**.

[0040] In one example, the 3D structure **10** may be comprised of an ionically conductive solid electrolyte using, for example, projection microstereolithography. The electrically conductive solid electrolyte has discrete, interpenetrating channels formed in it during the 3D printing process. The channels may be linear, but it will be appreciated that the channels will be non-linear for a 3D gyroid structure or most other 3D periodic or aperiodic structures. All the materials could be directly printed, and it is expected that this is likely to be a preferred implementation.

[0041] Subsequently, each of the channels **32** and **34** may be in-filled with active materials. For example, anode material may be filled into channel **32** and cathode material may be filled into channel **34**. Each of the active materials preferably has some conductive filler loaded into it before it is deposited in its respective channel **32** or **34** to improve the electrical conductivity of its associated anode or cathode material. Such conductive filler material may be Graphene, carbon nanotubes (CNTs), copper particles or wires, aluminum particles or wires, or carbon black. Again, a principal objective is to create a nonplanar current collector that is continuous and creates short electronic transport distances. Next, each anode and cathode material has portions thereof attached to a respective current collector using a conductive epoxy, such as was described in connection with material sheets **24** and **26** (i.e., current collectors) in FIG. **1**. An additional channel, represented by dashed line **36**, may be formed in the solid electrolyte **30** for the separator as well. The completed structure forms a battery which can then be tested. Ultimately, it is expected to be advantageous, from a manufacturing/cost standpoint, to directly pattern all of the current collector, active materials, conductive fillers, and separator/electrolyte directly with a 3D fabrication process.

[0042] Aside from tuning parameters in the 3D structures used in the design, these designs can be used with any combination of anode, cathode, electrolyte, separator, and current collector materials that are normally used in conventional battery designs. These 3D energy storage structures of the present disclosure are expected to be useful in both primary and secondary batteries, and could be applied in the construction of batteries for use in any application where power or energy density is a concern, either in terms of battery lifetime or energy storage capability on a single charge. Single charge storage capability is especially important for batteries used with mobile devices such as smartphones, tablets, laptops, MP3 players, gaming devices, GPS units, portable radios, power tools, home energy storage devices, grid storage devices or systems, or portable water purification units, just to name a few potential applications. The teachings provided herein are also expected to be important in helping to make batteries lighter for a given storage capacity, as compared to conventional battery designs. Minimizing the weight of the battery for a given level of power density is also expected to be especially important with applications involving many of the above listed devices, as well as with applications involving battery powered automotive vehicles, battery backup systems for use on aircraft, or even remotely controlled drones.

[0043] The present disclosure, in certain embodiments, makes use of geometries derived from triply periodic structures such as gyroids and Schwarz minimal surfaces, or other interpenetrating 3D structures, to achieve a significant improvement in power density over the previous conventional geometries at the same energy density and comparable feature (i.e., material thickness) sizes. A small number of examples of various types of periodic, 3D structures which may be used to form the 3D structure **10** are illustrated in FIGS. **4-12**, which show a Schwartz P surface (FIG. **4**), a Schwartz D surface (FIG. **5**), a Neovius surface (FIG. **6**), a N14 Surface (FIG. **7**), a N26 Surface (FIG. **8**), a N38 Surface (FIG. **9**), a Diamond surface (FIG. **10**), a Double Diamond

surface (FIG. **11**), and a Kagome lattice (FIG. **12**). The present disclosure may make use of any of the foregoing surfaces discussed herein or virtually any other surface provided at the following link:

[0044] http:www.susqu.edu/brake/evolver/examples/periodic/periodic.html.

[0045] The precise surface configuration could also be derived using shape or topology optimization to yield many different structures.

[0046] The various designs proposed in the present disclosure can be combined with improved battery materials to yield even further gains in battery performance over conventional designs using existing materials. It is expected that changes in material properties will affect the parameters determining the size and shape of the surfaces, but will not affect substantially the performance improvements obtained by using interpenetrating, periodic, 3D designs instead of conventional planar-based battery designs. It will be appreciated that interpenetration is a key feature, and it is desirable to maximize surface area without sacrificing active material.

[0047] The architectures of the present disclosure are expected to have particular utility with applications requiring portable power sources such as mobile phones, computing tablets and other portable electronic devices. The embodiments disclosed herein are also able to be charged more rapidly for a given level of energy than conventional batteries. The designs and teachings described herein may account for different capacities of the active materials. The designs and configurations discussed herein may also have different sizes and shape and amounts of active materials to boost overall battery capacity and efficiency.

[0048] The 3D energy storage architectures disclosed herein can also yield lighter or smaller batteries for a given quantity of energy storage, as compared to conventional planar or jelly roll layouts. This makes the various embodiments of the present disclosure especially valuable where weight is an important concern, such as with electronic devices used in military applications or with remotely controlled, battery powered land and air vehicles such as drones.

[0049] While various embodiments have been described, those skilled in the art will recognize modifications or variations which might be made without departing from the present disclosure. The examples illustrate the various embodiments and are not intended to limit the present disclosure. Therefore, the description and claims should be interpreted liberally with only such limitation as is necessary in view of the pertinent prior art.

What is claimed is:

1. An electrical energy storage apparatus, comprising:

an interpenetrating, three dimensional structure formed from an ionically conductive solid electrolyte material having a plurality of interpenetrating, non-planar channels, the interpenetrating, non-planar channels including:

a first plurality of channels filled with an anode material;

a second plurality of channels adjacent the first plurality of channels and interpenetrating the first plurality of channels, and filled with a cathode material;

a third plurality of channels adjacent, and interpenetrating, one of the first and second pluralities of channels and filled with a material to form a separator; and

said first, second and third channels forming a spatially dense, three dimensional structure;

a first non-flat current collector layer in communication with the first plurality of channels, and forming a first electrode; and

a second non-flat current collector layer in communication with the second non-planar channel and forming a second electrode.

2. The apparatus of claim **1**, wherein the anode material includes an electrically conductive filler material to improve electrical conductivity of the anode material.

3. The apparatus of claim **1**, wherein the cathode material includes an electrically conductive filler material to improve electrical conductivity of the cathode material.

4. The apparatus of claim **1**, wherein the three dimensional periodic structure comprises one of:

a gyroid;

a double gyroid;

a Schwartz surface;

kelvin foam;

octet truss, and

a kagome lattice;

a Neovius surface;

an N14 Surface;

an N26 Surface;

an N38 Surface;

a Diamond surface; and

a Double Diamond surface.

5. An electrical energy storage apparatus, comprising:

an interpenetrating, three dimensional periodic structure formed from an ionically conductive solid electrolyte material having a plurality of interpenetrating, non-planar channels, the plurality of interpenetrating, non-planar channels including:

a first plurality of channels of an anode material;

a second plurality of channels adjacent the first plurality of channels and interpenetrating the first plurality of channels, and being of a cathode material;

a third plurality of channels adjacent, and interpenetrating, one of the first and second pluralities of channels and being of a material to form a separator; and

a first current collector layer in communication with the first plurality of channels, and forming a first electrode;

a second current collector layer in communication with the second non-planar channel and forming a second electrode; and

wherein the interpenetrating, three dimensional periodic structure comprises one of:

a gyroid;

a double gyroid;

a Schwartz surface;

kelvin foam;

octet truss;

a kagome lattice;

a Neovius surface;

an N14 Surface;

an N26 Surface;

an N38 Surface;

a Diamond surface; and

a Double Diamond surface.

6. The apparatus of claim **5**, wherein each one of the first plurality of channels is filled with the anode material.

7. The apparatus of claim 6, wherein the anode material includes an electrically conductive filler material to improve electrical conductivity of the anode material.

8. The apparatus of claim 5, wherein each one of the second plurality of channels is filled with the cathode material.

9. The apparatus of claim 8, wherein the cathode material includes an electrically conductive filler material to improve electrical conductivity of the cathode material.

10. The apparatus of claim 5, wherein the first current collector layer comprises a non-flat current collector layer.

11. The apparatus of claim 5, wherein the second current collector layer comprises a non-flat current collector layer.

12. The apparatus of claim 5, wherein the interpenetrating, three dimensional periodic structure is formed using an additive manufacturing process.

13. A method for forming an electrical energy storage apparatus configured as a three dimensional structure, the method comprising:

forming an interpenetrating, three dimensional periodic structure having a first plurality of non-planar channels and a second plurality of non-planar channels in proximity to the first plurality of non-planar channels, the first and second pluralities of non-planar channels further being interpenetrating;

filling each one of the first plurality of non-planar channels with an anode material to form an anode;

filling each one of the second plurality of non-planar channels with a cathode material to form a cathode;

filling areas adjacent the first and second pluralities of non-planar channels with an electrolyte;

forming a first electrode to operate as a current collector, which is in electrical contact with portions of the anode material; and

forming a second electrode which is in electrical contact with portions of the cathode material.

14. The method of claim 13, further comprising adding electrically conductive filler material to the anode material before filling the first plurality of non-planar channels.

15. The method of claim 13, further comprising adding electrically conductive filler material to the cathode material before filling the second plurality of non-planar channels.

16. The method of claim 13, wherein the method forms an electrical energy storage device having at least one of the following configurations:

a gyroid;

a double gyroid;

a Schwartz surface;

kelvin foam;

an octet truss;

a kagome lattice;

a Neovius surface;

an N14 Surface;

an N26 Surface;

an N38 Surface;

a Diamond surface; and

a Double Diamond surface.

17. The method of claim 13, wherein the operation of forming the interpenetrating three dimensional structure comprises using a three dimensional printing process.

18. The method of claim 13, wherein the first electrode is formed by a first current collector layer of material, the first current collector layer of material being formed as a non-flat layer of material in interpenetrating engagement with the anode material.

19. The method of claim 13, wherein the second electrode is formed by a second current collector layer of material, the second current collector layer of material being formed as a non-flat layer of material in interpenetrating engagement with the cathode material.

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