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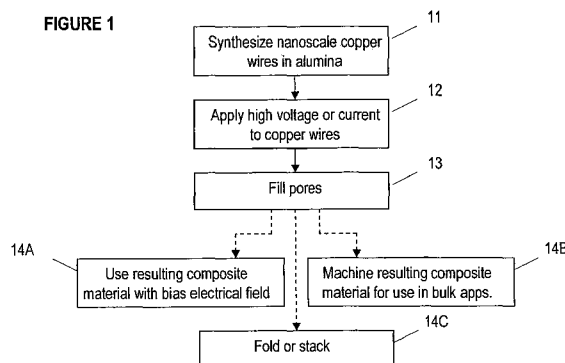
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(54) Title: HIGH DIELECTRIC PERMITTIVITY MATERIALS FROM COMPOSITES OF LOW DIMENSIONAL METALLIC SYSTEMS

FIGURE 1



(57) Abstract: Metal nanoparticles are assembled in interrupted metal strands or other structures of characteristic dimensions and orientation to generate a giant dielectric response through a modified GE effect. Careful selection and modification of the host material and synthesis also leads to low dielectric breakdown voltages. In addition, the high dielectric composite material is employed in material configurations that are more scalable for industrial and consumer applications.



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## HIGH DIELECTRIC PERMITTIVITY MATERIALS FROM COMPOSITES OF LOW DIMENSIONAL METALLIC SYSTEMS

### BACKGROUND

[001] Unless otherwise indicated herein, the approaches described in this section are  
5 not prior art to the claims in this application and are not admitted to be prior art by  
inclusion in this section.

[002] In 1965, Gor'kov and Eliashberg originally predicted that minute metallic particles  
should possess dramatically high polarizability, and thus a high dielectric constant,  
when small enough (i.e., nano-sized) such that their electronic energy levels are  
10 discrete. This effect is hereinafter referred to as the "GE effect." The symmetry of the  
spherical metallic particles, however, may induce a sufficient depolarization field from  
electrostatics to wash out the GE effect. This is explained in S. Strassler, et al.,  
"Comment on Gor'kov and Eliashberg's Result for the Polarizability of a Minute Metallic  
Particle," *Phys. Rev. B*, 6:2575 (1972), the contents of which are incorporated by  
15 reference herein.

[003] Further study on the GE effect was carried out in M. J. Rice, et al., "Gor'kov-  
Eliashberg Effect in One-Dimensional Metals?" *Phys. Review Lett.*, 29:113 (1972) (the  
"Rice publication"), the contents of which are incorporated by reference herein. In this  
publication, the researchers recognized that one-dimensional metals, such as mixed-  
20 valency planar complex compounds of platinum (Pt), might form interrupted metallic  
strands under sufficient conditions to manifest the GE effect. More recent research,  
published in S. K. Saha, "Observation of Giant Dielectric Constant in an Assembly of  
Ultrafine Ag Particles," *Phys. Rev. B*, 69:125416 (2004) (the "Saha publication"), the  
contents of which are incorporated by reference herein, has demonstrated the GE  
25 effect in interrupted metallic strands synthesized using modern techniques. In this and  
other studies including T. K. Kundu, et al., "Nanocomposites of Lead-Zirconate-Titanate  
Glass Ceramics and Metallic Silver," *Appl. Phys. Lett.*, 67:2732 (1995) (the "Kundu  
publication) and B. Roy, et al., "High Dielectric Permittivity in Glass-Ceramic Metal  
Nanocomposites," *J. Mater. Res.*, 8:1206 (1993), the contents of which are

incorporated by reference herein, researchers have demonstrated giant dielectric responses (on other order of  $\epsilon \sim 10^{10}$ ) in small-scale assemblies of ultrafine metal particles (i.e., metal nanoparticles) under external electrical bias, disordered metal/semiconductor particles without bias, and at various temperatures and frequencies.

## SUMMARY

[004] In accordance with at least some embodiments of the present disclosure, a method of producing a high dielectric permittivity composite material is disclosed. The method includes selecting alumina as a host material, synthesizing nanoscale copper wires in the host material, applying a current in the range of 100  $\mu$ A to 10 mA to produce copper atom islands in interrupted strands, and filling pores in the host material that are not filled with the copper wires.

[005] In accordance with at least some other embodiments of the present disclosure, a large scale structure that is at least 1 mm thick is also disclosed. The large scale structure includes multiple layers of composite material having high dielectric constants due to the GE effect.

## BRIEF DESCRIPTION OF THE DRAWINGS

[006] Figure 1 is a flow diagram of an illustrative embodiment of a process for producing a composite material having a high dielectric constant;

[007] Figure 2 is a flow diagram of an illustrative embodiment of a process for producing a composite material having a high dielectric constant; and

[008] Figures 3A and 3B illustrate two examples of a large scale structure that contains high dielectric permittivity materials.

## DETAILED DESCRIPTION

[009] In the following detailed description, reference is made to the accompanying drawings, which form a part hereof. In the drawings, similar symbols typically identify

similar components, unless context dictates otherwise. The illustrative embodiments described in the detailed description, drawings, and claims are not meant to be limiting. Other embodiments may be utilized, and other changes may be made, without departing from the spirit or scope of the subject matter presented herein. It will be readily understood that the aspects of the present disclosure, as generally described herein, and illustrated in the Figures, can be arranged, substituted, combined, separated, and designed in a wide variety of different configurations, all of which are explicitly contemplated herein.

[0010] This disclosure is drawn, *inter alia*, to the synthesis, production, and use of new dielectric composites of low-dimensional metallic or metal-like particles and molecular templates that guide their synthesis. These particles are assembled in interrupted metal strands or other structures of characteristic dimensions and orientation to generate a giant dielectric response through a modified GE effect. Careful modification of the composite host material also leads to low dielectric breakdown voltages.

Materials with high dielectric constants and/or improved voltage breakdown characteristics are useful as they help enable advances in supercapacitor applications and technologies.

[0011] One example of a high dielectric permittivity material is a polycarbonate membrane having channels that are filled with ultrafine particles of silver in close physical proximity to each other. This is the material described in the Saha publication. These form interrupted strand configurations similar to a linear strand of beads, with each ultrafine particle of silver as a bead. The silver particles have an effective diameter on the order of nanometers and overall strand lengths of  $\sim 50 \mu\text{m}$ . An overall composite  $50 \mu\text{m}$  thick is estimated to have a dielectric constant of  $10^{10}$  along the strand axis. An electrical field bias of 0.05 volts is required to achieve a capacitive state.

[0012] Other examples of a high dielectric permittivity material is a nanocomposite of PZT glass ceramics and metallic silver that is described in the Kundu publication, which exhibits a dielectric constant of 300-1000 at 300 K, and silver nanowire assemblies in

mica described in P. K. Mukherjee, et al., "Growth of Silver Nanowires Using Mica Structure as a Template and Ultrahigh Dielectric Permittivity of Nanocomposite," *J. Mater. Res.*, 17:3127 (2002), the contents of which are incorporated by reference herein, which exhibits a dielectric constant of around  $10^7$ . In certain examples where  
5 the particles are spherical, an applied electrical field bias is necessary to physically distort the particles and break the spherical symmetry to induce a large dielectric response.

[0013] Silver is used in many of the examples because of ease of synthesis and the high yield of pore-filling reaction. However, polypyrrole nanorods as disordered metals  
10 may also be used, as shown in S. K. Saha, "One-Dimensional Organic Giant Dielectrics," *App. Phys. Lett.*, 89:043117 (2006), the contents of which are incorporated by reference herein.

[0014] As used herein, an interrupted metallic strand is an aggregate of metal particles joined by physical proximity in a lattice, though break junctions, insulating junctions, or  
15 other means. These strands may be metal particles in a linear formation interrupted by endogeneous lattice defects as described in the Rice publication. The linear formations between the junctions may be conceived as deep potential wells and modeled as a 1D sequence of "particle-in-a-box" potentials. The model estimates a dielectric constant on the order of the particle lengths, and when integrated over the ensembles of strands,  
20 gives the very high values of dielectric constants shown by previous researchers. In some cases, the composite material may be considered as a dual lattice exhibiting a Maxwell-Wagner space charge mechanism, particularly at high frequencies and temperatures.

[0015] The present disclosure extends the range of systems that benefit from the GE  
25 effect. Extensions of both active metallic materials and host materials contained in such systems are contemplated. This disclosure also introduces novel components for the production of these materials, such as for void-filling, and large structure construction. Even in non-optimal cases, these materials would enhance dielectric constant by orders of magnitude. In addition, material configurations that are more

appropriate for large scale applications are described. These material configurations are more scalable for industrial and consumer applications than the single, thin membranes taught in the prior art and minimize the open pores that do not contain nanowires because such open pores become air filled gaps that contribute to electrical breakdown (e.g., by arcing).

[0016] The composite materials and the material configurations set forth in this disclosure include one or more of the following features:

- 1. Particle assemblies of the correct size. Metal or metal-like material particle assemblies are synthesized in host materials near the characteristic length for the particular material so that the GE effect is observed at room temperature or other appropriate operating temperature. The materials may include one or more of Cu, Mg, Au, Zn, Cd, Al, Mg, K, or metal-like materials, such as conducting polymers or conjugated molecular systems.
- 2. Host materials and chemistries. A porous host material with a pore diameter suitable to template nanoscale assemblies exhibiting quantum energy states (order of 100 nm or less) is selected. The selected host material facilitates chemistry for interrupted strand synthesis of precursors. Host materials may include one of the following classes: porous materials such as alumina, polycarbonate/other organic polymers, zeolites, molecular sieves, other mesoporous materials, or template materials such as mica or glass ceramics. Chemistries may include electro-deposition, pyrolytic, other redox and synthesis reactions. Any voltaic synthesis may use the host as an anode.
- 3. Application of a post-synthesis step. An optional post-synthesis step is applied to produce the actual interrupted metal strand or final nanostructures. The post-synthesis step may include sintering, current-induced melting (as with silver nanowire conversion to ultrafine particles), or other physio-chemical methods that convert wires or homogeneous structures from the particle assemblies from #1 above into interrupted metal strands.

- 4. Pore blocking. A supplementary sequence of reactions is carried out to block the remaining pores and prevent these pores from becoming air filled gaps in the final structure. These reactions may include follow-on deposition reactions that go to completion far better than the main reaction. For example, in the case where copper (Cu) wires in alumina are created to exhibit the GE effect, the remaining porous space may be filled with polyester.
  - 5. Post-processing for large structures. Further processing of the composite material is carried out to create an application-relevant structure. This may include folding of filled membranes to create a thick structure, compacting of zeolites filled with nanowires into dense materials, or machining of filled alumina into particles for further use.
  - 6. Use as capacitors. The composite material may be used as high dielectric materials in applications requiring capacitors with or without favorable high voltage characteristics, in some cases, with an applied voltage bias.
  - 7. Use of applied voltage bias. In some embodiments, e.g., in ultrafine assemblies of metals in spherical configurations, a voltage bias is applied to the composite material when it is used in a capacitor structure. The voltage bias may result from an extrinsic field or an intrinsic field. The voltage bias may not be necessary with nanorods or other non-spherical particles.
- The characteristic number of metal atoms for a structure to exhibit the GE effect is based on the spatial length of the "particle-in-a-box" potential at the appropriate temperature. Table 1 below shows the characteristic number of metal atoms for various metals at room temperature.

TABLE 1

| Element       | Ionic Radius (pm) | Number of Atoms |
|---------------|-------------------|-----------------|
| Platinum (Pt) | 150               | 267             |

|                |      |     |
|----------------|------|-----|
| Copper (Cu)    | 77   | 519 |
| Silver (Ag)    | 94   | 425 |
| Gold (Au)      | 85   | 471 |
| Zinc (Zn)      | 74   | 540 |
| Cadmium (Cd)   | 95   | 421 |
| Beryllium (Be) | 45   | 889 |
| Magnesium (Mg) | 72   | 556 |
| Aluminum (Al)  | 53.5 | 748 |
| Potassium (K)  | 138  | 290 |
| Sodium (Na)    | 102  | 392 |

[0017] The following are some embodiments that include one or more features of the present disclosure.

[0018] Example 1. Copper in alumina and machined alumina. Figure 1 is a flow diagram of an illustrative embodiment of a process for producing a composite material having a high dielectric constant. Nanoscale copper wires are first synthesized in alumina as a host via electro-deposition with the alumina as the anode (Block 11). The synthesis is described in further detail in T. Gao, et al., "Electrochemical Synthesis of Copper Nanowires," *J. Phys.: Condens. Matter*, 14:255 (2002), the contents of which are incorporated by reference herein. At Block 12, a high voltage/current is applied to the nanoscale copper wires in alumina. A current between 100  $\mu$ A to 10 mA is carefully selected to create assemblies of ~ 500 Cu atom islands in interrupted strands. A low temperature polymerization of polyester then follows to fill those pores that do not go to synthetic completion (i.e., are not filled with the copper nanowires) (Block 13). The resulting composite material may be used with a bias electrical field to generate a large dielectric response along the strand axis (Block 14A). The resulting composite material may also be machined into powder and used in bulk applications (Block 14B). Though the assemblies would become randomly oriented, it is expected that a sufficient number would be oriented in a particular direction of an applied field to account for a strong

enhancement of the overall dielectric constant. In addition, the resulting composite material may be folded or stacked to create a large-scale structure (Block 14C).

[0019] Example 2. Potassium in zeolites. Figure 2 is a flow diagram of an illustrative embodiment of a process for producing a composite material having a high dielectric constant. Nanoscale metals may be synthesized in structures such as zeolites, e.g., 1D channel zeolites and some 2D and 3D channel zeolites that have non-interconnected channels. 1D channel zeolites may include Zeolite L, Zeolite-Linde-Type-L (LTL), AIPO-31 (ATO) zeolite, roggianite (-RON) zeolite, EU-1 (EUO) zeolite, RUB-3 (RTE) zeolite, and other 1D channel zeolites disclosed in Xu, R., et al., *Chemistry of Zeolites and Related Porous Materials: Synthesis and Structure*, Wiley-Interscience, pp. 44 – 46 (2007).

[0020] At Block 21, Zeolite L is selected as the host material. Then, at Block 22, conventional Davy electrolysis of KOH is carried out with Zeolite L to produce potassium nanowires in Zeolite L. At block 23, Zeolite L that is filled with potassium nanowires is compacted into dense materials for use in an application-relevant structure. By choosing a zeolite with appropriate channel length, such as Zeolite L (which can be grown to crystal lengths of 20 to 7000 nm), potassium nanowires may be grown in each channel exactly matching the optical distance (~ 80 nm) to manifest the GE effect. Zeolites as a host have the advantage of creating an ensemble of single length nanowire segments matched precisely to the “particle-in-a-box” length, instead of relying on kinetically or thermodynamically controlled reactions in extended mesoporous channels. Though Zeolite L has strictly linear channels, each particle will be oriented randomly in space (zeolites with nonlinear channels will be oriented equivalently in the aggregate), leaving only a fraction oriented parallel to any applied field. By simple geometric integration of the dielectric vector, the enhancement of dielectric constant is still expected to be high order, only, at most, a few orders of magnitude lower than the  $\epsilon \sim 10^{10}$  of an oriented system. Additionally, because each wire represents a single “particle-in-a-box” potential, applications need no applied bias field.

[0021] Example 3. Though silver ultrafine particles have been created in thin (50  $\mu\text{m}$ ) polycarbonate membranes to create high dielectric compounds, overall scale up from these structures has not been contemplated. However, polycarbonate or other flexible membranes could form the basis of a roll-to-roll manufacturing process, with sheets that are folded together, cut and stacked, or rolled to create thick structures. A 10 cm thick structure includes approximately 2000 sheets of membrane that are stacked or folded. Rolled structures easily scale to consumer power cells (similar to consumer battery cells that are constructed from rolled electrodes). Figures 3A and 3B illustrate two examples of a large scale structure that contains high dielectric permittivity materials.

In these examples, large scale structure 31 includes multiple stacked sheets of polycarbonate membrane 32 containing silver nanoparticles 33 (Figure 3A) or multiple folded sheet of polycarbonate membrane 32 containing silver nanoparticles 33 (Figure 3B). In other embodiments, large scale structure 31 may include multiple folded or stacked sheets of other materials having high dielectric constants due to the GE effect.

[0022] Example 4. According to the Rice publication, nanorods of polypyrrole have been synthesized using low temperature pyrolysis in alumina and shown to exhibit giant dielectric effects. The mechanism is posited to be disordered metal phases interrupted by semiconductor phases to create interrupted strand structures. Other work, published in J. I. Lee, et al., "Highly Aligned Ultrahigh Density Arrays of Conducting Polymer Nanorods Using Block Copolymer Templates," *Nano Lett.*, 8:2315 (2008) (the "Lee publication"), the contents of which are incorporated by reference herein, teaches electrodepositing of polypyrrole on indium tin oxide (ITO) to create ultrahigh density vertical arrays of highly conductive rods (though they have not been tested for capacitive function). According to one or more embodiments of this disclosure, many other conducting polymers, ranging from polyaniline to more exotic specialty polymers may be synthesized in host materials, replacing metals used in the Rice and Lee publications. In addition, control of dopant levels through known chemical techniques, such as kinetic control, allows good control of interrupted strand dimensions and defect density. The low weight of these polymers make them ideal for creating systems with a low weight-to-performance ratio.

[0023] Capacitor system with high effective permittivity enables multibillion dollar energy markets ranging from portable electronics to automotive to large power systems. The materials set forth in the present disclosure would enable applications across these markets, particularly those requiring very high dielectric constants.

- 5 [0024] Industrial and academic efforts have produced high dielectric materials, but each with considerable associated difficulties. Many ceramic high dielectric materials for parallel plate capacitors suffer from low breakdown levels because of material structural defects. Other materials with better processing characteristics have insufficient dielectric properties for large scale use (e.g., dielectric constants in the tens, not  
10 hundreds). Though researchers have contemplated nanodielectrics for industrial applications as published in C. Yang, et al., "The Future of Nanodielectrics in the Electrical Power Industry," *IEEE Trans. Dielec. and Elec. Insul.*, 11:797 (2004), the contents of which are incorporated by reference herein, little work has been performed to target structures and systems that are appropriate for non-laboratory applications.
- 15 [0025] One exception may be the company, EESor. EESor is the assignee of U.S. Patent Nos. 7,033,406 and 7,466,536, which are directed to low void and low defect BaTiO<sub>3</sub> structures. BaTiO<sub>3</sub> possesses abnormally large dielectric constants but voids and defects from traditional syntheses lead to poor electric breakdown robustness. EESor claims to have solved this problem with low-temperature and kinetically labile  
20 synthetic routes. However, considerable skepticism remains about the commercial viability and scalability of their product. In addition, even in the EESor materials, dielectric constants of  $\epsilon > 10^4$  are unlikely in production.

[0026] With respect to the use of substantially any plural and/or singular terms herein, those having skill in the art can translate from the plural to the singular and/or from the  
25 singular to the plural as is appropriate to the context and/or application. The various singular/plural permutations may be expressly set forth herein for sake of clarity.

[0027] It will be understood by those within the art that, in general, terms used herein, and especially in the appended claims (e.g., bodies of the appended claims) are

generally intended as "open" terms (e.g., the term "including" should be interpreted as "including but not limited to," the term "having" should be interpreted as "having at least," the term "includes" should be interpreted as "includes but is not limited to," etc.).

It will be further understood by those within the art that if a specific number of an

5 introduced claim recitation is intended, such an intent will be explicitly recited in the claim, and in the absence of such recitation no such intent is present. For example, as an aid to understanding, the following appended claims may contain usage of the introductory phrases "at least one" and "one or more" to introduce claim recitations.

However, the use of such phrases should not be construed to imply that the

10 introduction of a claim recitation by the indefinite articles "a" or "an" limits any particular claim containing such introduced claim recitation to inventions containing only one such recitation, even when the same claim includes the introductory phrases "one or more" or "at least one" and indefinite articles such as "a" or "an" (e.g., "a" and/or "an" should typically be interpreted to mean "at least one" or "one or more"); the same holds true for

15 the use of definite articles used to introduce claim recitations. In addition, even if a specific number of an introduced claim recitation *is* explicitly recited, those skilled in the art will recognize that such recitation should typically be interpreted to mean *at least* the recited number (e.g., the bare recitation of "two recitations," without other modifiers,

typically means *at least* two recitations, or *two or more* recitations). Furthermore, in

20 those instances where a convention analogous to "at least one of A, B, and C, etc." is used, in general such a construction is intended in the sense one having skill in the art would understand the convention (e.g., "a system having at least one of A, B, and C" would include but not be limited to systems that have A alone, B alone, C alone, A and B together, A and C together, B and C together, and/or A, B, and C together, etc.). In

25 those instances where a convention analogous to "at least one of A, B, or C, etc." is used, in general such a construction is intended in the sense one having skill in the art would understand the convention (e.g., "a system having at least one of A, B, or C" would include but not be limited to systems that have A alone, B alone, C alone, A and

30 B together, A and C together, B and C together, and/or A, B, and C together, etc.). It will be further understood by those within the art that virtually any disjunctive word

and/or phrase presenting two or more alternative terms, whether in the description, claims, or drawings, should be understood to contemplate the possibilities of including one of the terms, either of the terms, or both terms. For example, the phrase "A or B" will be understood to include the possibilities of "A" or "B" or "A and B."

5 [0028] From the foregoing, it will be appreciated that various embodiments of the present disclosure have been described herein for purposes of illustration, and that various modifications may be made without departing from the scope and spirit of the present disclosure. Accordingly, the various embodiments disclosed herein are not  
10 intended to be limiting, with the true scope and spirit being indicated by the following claims.

**Claims**

1. A method of producing a high dielectric permittivity composite material, comprising:

5 selecting a host material with nano- or micro-scale pores or mesoporous channels;

synthesizing conductive material in the host material channels to form interrupted strands; and

10 filling the pores in the host material that are not filled with the conductive material.

2. The method of claim 1, wherein the host material is alumina, the conductive material copper and the pore filling material polyester, and a current in the range of 100  $\mu$ A to 10 mA is applied to produce copper atom islands in interrupted strands.

15 3. The method of claim 2, wherein the filling comprises a low temperature polymerization of polyester.

20 4. The method of claim 2, wherein the nanoscale copper wires are synthesized via electro-deposition with the alumina as the anode.

25 5. The method of claim 2, wherein the filling comprises a low temperature polymerization of polyester and the nanoscale copper wires are synthesized via electro-deposition with the alumina as the anode.

6. The method of claim 2, further comprising:  
applying a bias electrical field in a manner to generate a large dielectric response along an axis of the host material filled with the copper wires.

7. The method of claim 2, further comprising:  
machining the host material filled with the copper wires.

8. The method of claim 2, further comprising:  
5 stacking multiple sheets of the host material filled with the copper wires.

9. A method of producing a high dielectric permittivity composite material  
comprising:

10 selecting a zeolite as a host material; and  
synthesizing conductive material in the host material.

10. The method of claim 9, further comprising:  
compacting the host material containing the conductive material.

15 11. The method of claim 10, wherein the host material has channel lengths of 20 to  
7000 nm.

12. The method of claim 11, wherein the host material containing the conductive  
material has a dielectric constant that is at least  $10^8$ .

20 13. The method of 9, wherein the host material is Zeolite L and the conductive  
material is potassium.

14 The method of claim 13, wherein potassium nanowires are synthesized in Zeolite  
25 L by the Davy electrolysis of KOH.

15. The method of claim 14, wherein the potassium nanowires are each formed to  
have an optical distance of about 80 nm.

16. A high dielectric permittivity composite material having a dielectric constant of at least  $10^8$  comprising potassium nanowires in Zeolite L.
17. A large scale structure that is at least 1 mm thick comprising multiple layers of composite material having high dielectric constants due to the GE effect.
18. The large scale structure of claim 17, wherein the multiple layers are formed by folding one or more sheets of the composite material.
19. The large scale structure of claim 17, wherein the multiple layers are formed by stacking one or more sheets of the composite material.
20. The large scale structure of claim 17, wherein the composite material includes polycarbonate membranes containing silver nanoparticles.
21. The large scale structure of claim 17, wherein the composite material includes alumina containing copper nanoparticles.
22. The large scale structure of claim 17, wherein the composite material includes alumina containing copper nanoparticles and polyester.

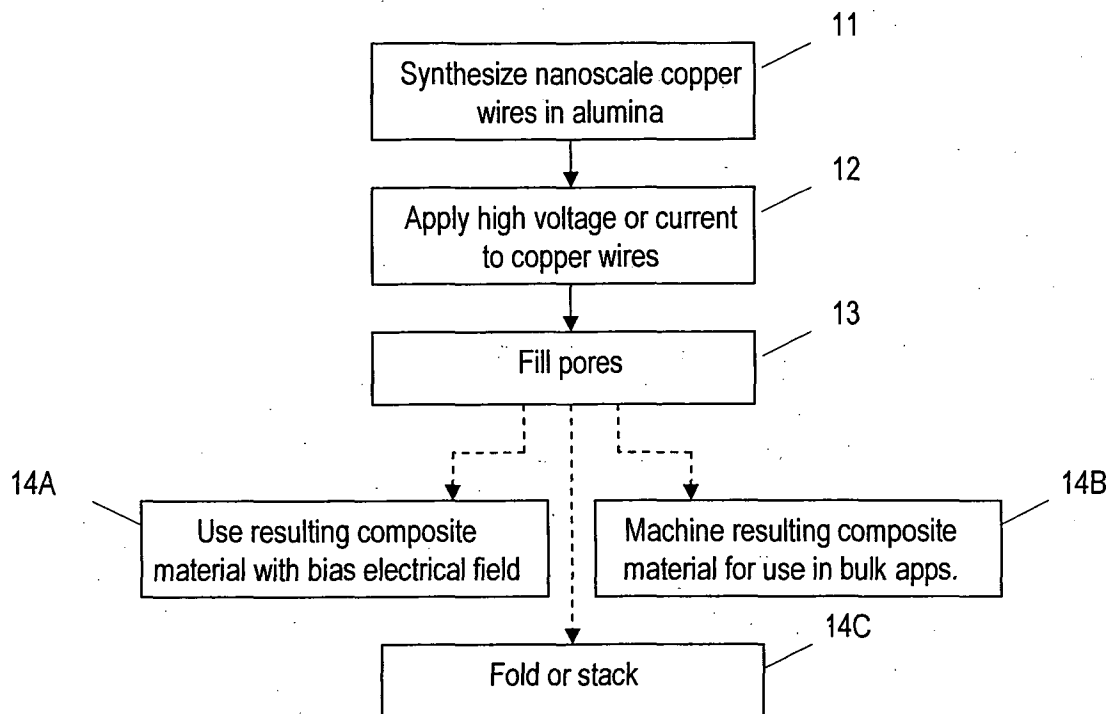


FIGURE 1

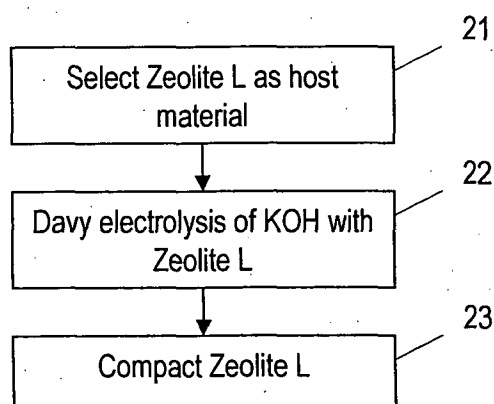


FIGURE 2

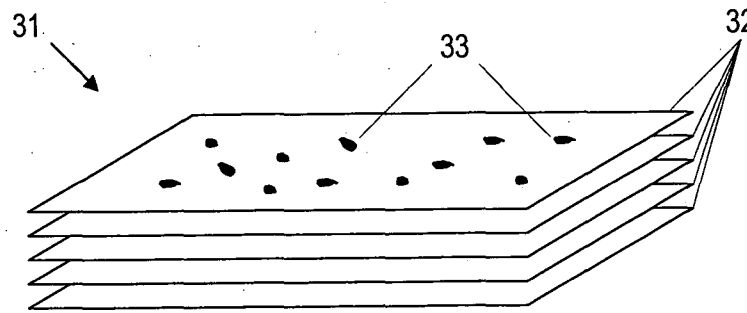


FIGURE 3A

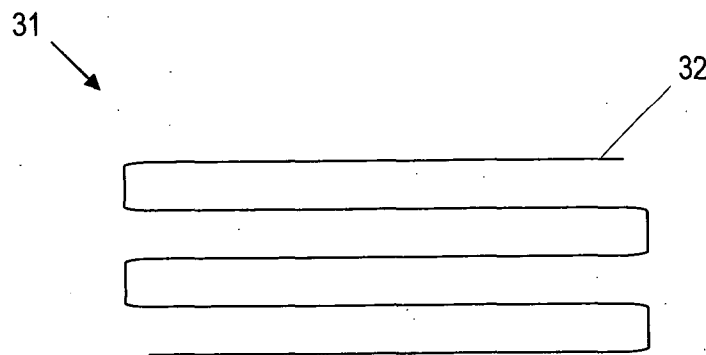


FIGURE 3B

# INTERNATIONAL SEARCH REPORT

International application No.

PCT/SG2010/000191

## A. CLASSIFICATION OF SUBJECT MATTER

Int. Cl. **H01G 4/08** (2006.01) **C01B 39/02** (2006.01) **B82B 3/00** (2006.01) **H01G 4/22** (2006.01)

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)

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)

WPI, EPODOC, HCA, INSPEC – Keywords: dielectric, GE effect, Gorkov, Eliashberg, capacitance, capacitor, farad, zeolite, molecular sieve, nanoporous, microporous, aluminosilicate, sponge, alumina, deposit, apply, coat, electroplate, electrodeposit, current, electrolysis, pvd, cvd, nanoparticles, ultrafine, island, interrupted, strand, nanowire, nanorod, isolate, nanotube, nanowhisker, copper, cu, silver, ag, potassium, k, polymer, polyester, polycarbonate, and like terms.

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category* | Citation of document, with indication, where appropriate, of the relevant passages                                                                                                                                            | Relevant to claim No. |
|-----------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------|
| A         | US 2005/0136608 A1 (MOSLEY) 23 June 2005<br>See paragraphs [0021], [0022], [0030]                                                                                                                                             |                       |
| A         | US 2006/0108687 A1 (BOYANOV et al.) 25 May 2006<br>See abstract                                                                                                                                                               |                       |
| A         | Takano, T, et al., 'μSR study on ferromagnetism of potassium clusters in aluminosilicate zeolite LTA' Physica B, 2003, vol. 326, pages 550-555<br>See abstract                                                                |                       |
| A         | Saha, S, 'Observation of giant dielectric constant in an assembly of ultrafine Ag particles', Physical Review. B; Condensed Matter and Materials Physics, 2004, vol. 69(12), pages 125416-1 to 125416-4<br>See whole document |                       |



Further documents are listed in the continuation of Box C



See patent family annex

|                                                                                                                                                                         |                                                                                                                                                                                                                                                  |
|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| * Special categories of cited documents:                                                                                                                                |                                                                                                                                                                                                                                                  |
| "A" document defining the general state of the art which is not considered to be of particular relevance                                                                | "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention                                              |
| "E" earlier application or patent but published on or after the international filing date                                                                               | "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone                                                                     |
| "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) | "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art |
| "O" document referring to an oral disclosure, use, exhibition or other means                                                                                            | "&" document member of the same patent family                                                                                                                                                                                                    |
| "P" document published prior to the international filing date but later than the priority date claimed                                                                  |                                                                                                                                                                                                                                                  |

Date of the actual completion of the international search  
21 July 2010

Date of mailing of the international search report  
22 JUL 2010

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# INTERNATIONAL SEARCH REPORT

International application No.

PCT/SG2010/000191

## Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:  
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
3. ☐ Claims Nos.:  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a)

## Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

As reasoned in Supplemental Box 1.

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☒ As all searchable claims could be searched without effort justifying additional fees, this Authority did not invite payment of additional fees.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

### Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.
- ☐ The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.
- ☐ No protest accompanied the payment of additional search fees.

# INTERNATIONAL SEARCH REPORT

International application No.

PCT/SG2010/000191

## Supplemental Box 1

(To be used when the space in any of Boxes I to IV is not sufficient)

### Continuation of Box No: III

This International Application does not comply with the requirements of unity of invention because it does not relate to one invention or to a group of inventions so linked as to form a single general inventive concept.

In assessing whether there is more than one invention claimed, I have given consideration to those features which can be considered to potentially distinguish the claimed combination of features from the prior art. Where different claims have different distinguishing features they define different inventions.

This International Searching Authority has found that there are different inventions as follows:

- Claims 1-8 relate to a method of producing a high dielectric permittivity composite material, the method comprising: selecting a host material with nano- or micro- pores or mesoporous channels; synthesising conductive material in the host material channels to form interrupted strands; and filling the pores in the host material that are not filled with the conductive material. It is considered that: synthesising interrupted strands of conductive material; and filling pores that are not filled with conductive material comprises a first distinguishing feature.
- Claims 9-16 relate to a method of producing a high dielectric permittivity composite material, the method comprising: selecting a zeolite as a host material; and synthesising conductive material in the host material. It is considered that synthesising conductive material in a zeolite comprises a second distinguishing feature.
- Claims 17-22 relate to a large scale structure that is at least 1mm thick comprising multiple layers of composite material having high dielectric constants due to the GE effect. It is considered that multiple layers of material having high dielectric constants due to the GE effect comprises a third distinguishing feature.

PCT Rule 13.2, first sentence, states that unity of invention is only fulfilled when there is a technical relationship among the claimed inventions involving one or more of the same or corresponding special technical features. PCT Rule 13.2, second sentence, defines a special technical feature as a feature which makes a contribution over the prior art.

The only feature common to all of the claims is a high dielectric permittivity composite material. However this common feature is generic in the art. This means that the common feature can not constitute a special technical feature within the meaning of PCT Rule 13.2, second sentence, since it makes no contribution over the prior art.

Because the common feature does not satisfy the requirement for being a special technical feature it follows that it cannot provide the necessary technical relationship between the identified inventions. Therefore the claims do not satisfy the requirement of unity of invention *a posteriori*.

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

**PCT/SG2010/000191**

This Annex lists the known "A" publication level patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

| Patent Document Cited in<br>Search Report                                                                                 |            |    |            | Patent Family Member |            |    |            |
|---------------------------------------------------------------------------------------------------------------------------|------------|----|------------|----------------------|------------|----|------------|
| US                                                                                                                        | 2005136608 | US | 7019346    | US                   | 2005136609 | US | 7242073    |
|                                                                                                                           |            | US | 2006115951 | US                   | 7288460    |    |            |
| US                                                                                                                        | 2006108687 | CN | 1808698    | US                   | 7303989    | WO | 2006058009 |
| Due to data integration issues this family listing may not include 10 digit Australian applications filed since May 2001. |            |    |            |                      |            |    |            |
| END OF ANNEX                                                                                                              |            |    |            |                      |            |    |            |