In a method of making a thermal interface material, functionalized graphene sheets are dispersed in a liquid. The liquid is removed through a filter so as to form a filtration cake of aligned functionalized graphene sheets, substantially all of which are parallel to a common plane. At least one block of aligned functionalized graphene sheets is cut from the filtration cake. The block includes a first end face and an oppositely-disposed second end face that are parallel to each other and to which substantially all of the functionalized graphene sheets are transverse. The block also includes two oppositely-disposed sides to which substantially all of the functionalized graphene sheets are parallel. A first layer of a thermally conductive substance is applied to the first end face and a second layer of the thermally conductive substance is applied to the second end face of the block.

19 Claims, 2 Drawing Sheets
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Three-Dimensional Vertically Aligned Functionalized Multilayer Graphene

Summary of the Invention

The disadvantages of the prior art are overcome by the present invention, which, in one aspect, is a method of making a thermal interface material, in which functionalized graphene sheets are dispersed in a liquid. The liquid is removed through a filter so as to form a filtration cake of aligned functionalized graphene sheets, substantially all of which are parallel to a common plane. At least one block of aligned functionalized graphene sheets is cut from the filtration cake. The block includes a first end face and an oppositely disposed second end face that are parallel to each other and to which substantially all of the functionalized graphene sheets are transverse. The block also includes two oppositely disposed sides to which substantially all of the functionalized graphene sheets are parallel. A first stabilizing layer is applied to the first end face and a second stabilizing layer is applied to the second end face. The first stabilizing layer is placed to be in thermal communication with the heat source and the second stabilizing layer is placed to be in thermal communication with the heat sink.

In yet another aspect, the invention is a thermal interface material, that includes a block of a plurality of functionalized graphene sheets that are substantially all aligned so as to be parallel to a common plane. The block has a first end face that is transverse to the common plane and a second end face that is transverse to the common plane. A first layer of a thermally conductive substance is applied to the first end face and a second layer of the thermally conductive substance is applied to the second end face. A first silicon layer is applied to the first layer of the thermally conductive substance and a second silicon layer is applied to the second layer of the thermally conductive substance.

These and other aspects of the invention will become apparent from the following description of the preferred embodiments taken in conjunction with the following drawings. As would be obvious to one skilled in the art, many variations and modifications of the invention may be effected without departing from the spirit and scope of the novel concepts of the disclosure.

Detailed Description of the Invention

A preferred embodiment of the invention is now described in detail. Referring to the drawings, like numbers indicate like parts throughout the views. Unless otherwise specifically indicated in the disclosure that follows, the drawings are not necessarily drawn to scale. As used in the description herein and throughout the claims, the following terms take the meanings explicitly associated herein, unless the context clearly dictates otherwise: the meaning of “a,” “an,” and “the” includes plural reference, the meaning of “in” includes “in” and “on.”

As shown in FIGS. 1A-1K, one method of making a vertically aligned functionalized multilayer graphene block 130 begins with making a dispersion 110 by dispersing a plurality of graphene sheets 114 in a liquid 112 such as water. The liquid 112 is removed from the dispersion through a filter 116, such as by applying a vacuum to the filter 116. In one embodiment, the filter 116 includes an aluminum oxide filtration paper. As the liquid 112 is removed, the graphene sheets 114 will lay down in an orientation in which they are substantially aligned so as to be parallel to a common plane. Eventually, substantially all of the liquid 112 is removed, leaving a cake 120 of graphene sheets 114 that are aligned with each other.
The cake 120 may then be cut along predetermined lines 122, thereby generating a plurality of blocks 130 of aligned graphene.

Each block includes a first end face 132, a second end face 133 that is opposite from the first end face 132, a third end face 138, an opposite fourth end face 139, a top side 134 and an opposite bottom side 136. Substantially all of the graphene sheets 114 are aligned parallel to the top side 134 and are transverse to the first end face 132.

A plurality of the blocks 130 are rotated so that the first end face 132 is facing upwardly. The blocks 130 are then drawn together so as to form a common top surface 140 and an opposite bottom surface 142. A layer of a thermally conductive substance 144, such as indium, is applied to both the top surface 140 and the bottom surface 142 and then a layer of silicon 146 is applied to each layer of the thermally conductive substance 144, resulting in the formation of a thermal interface material unit 148.

As shown in FIG. 2, the thermally conductive substance 144 adheres to the graphene sheet blocks 130 and the silicon layer adheres to the thermally conductive substance 144. The graphene sheets may also be functionalized with functionalizing molecules 158 so as to overcome forces that might interfere with their alignment. The graphene may be functionalized, for example, with functional groups such as conjugated carbon-carbon, carbon-hydroxyls, and carboxyls.

In one embodiment, as shown in FIG. 3, the thermal interface material unit 148 can be thermally coupled to a heat source 156 (such as an integrated circuit) and a heat sink 158. Because of the superior conductivity of graphene along its planar axis, heat transfer from the heat source 156 to the heat sink 158 through the thermal interface material unit 148 is highly efficient.

In one experimental embodiment, functionalized multilayer graphene (fMG) was synthesized and 0.8 g of the fMG sheets were dispersed in 1 L of deionized water. The dispersion was vacuum-filtrated with a 47 mm vacuum filtration system equipped with an anodic aluminum oxide (AAO) filtration paper (0.1 µm pore size, Anodisc 47, Whatman International Ltd.). The obtained filtration cake was washed by diionized water, removed from filtration paper, and dried at 105 °C for 3 hours. In order to obtain a high thermally conductive array, reduction-free thermally conductive surface functionalized multilayer graphene sheets (functionalized multilayer graphene) were aligned. Functionalized multilayer graphene sheets were prepared in a moderate oxidation environment (mixed sulfuric and nitric acid) with aid of sonication. The thickness of the functionalized multilayer graphene sheets was in the range of between 2 nm and 10 nm, with an average thickness of 7.35 nm.

Graphene is susceptible to van der Waals forces and tends to be recumbent on substrates. If aligned and densely packed in an array, multilayer graphene sheets can be self-supporting, conveniently rearranged, and vertically assembled between solid surfaces thereafter. In order to obtain a high thermally conductive array, reduction-free thermally conductive surface functionalized multilayer graphene sheets (functionalized multilayer graphene) were aligned. The thickness of the functionalized multilayer graphene sheets was in the range of between 2 nm and 10 nm, with an average thickness of 7.35 nm.

Silicon wafers (1 mm by 1 mm) were coated with melted pure indium at a temperature of 180 °C. The thickness of the indium coating was about 10 µm after careful polishing at room temperature. A-functionalized multilayer graphene samples were sliced and sandwiched between thus-prepared Silicon wafers with an orientation of functionalized multi-layer graphene perpendicular to the contacted Silicone surfaces, corresponding to vertically aligned-functionalized multilayer graphene thermal interface material assemblies. The sandwiched samples were clamped with small pressure (about 0.02 MPa) and placed in a convection oven at 200 °C for 20 minutes and then cooled to room temperature.

The above described embodiments, while including the preferred embodiment and the best mode of the invention known to the inventor at the time of filing, are given as illustrative examples only. It will be readily appreciated that many deviations may be made from the specific embodiments disclosed in this specification without departing from the spirit and scope of the invention. Accordingly, the scope of the invention is to be determined by the claims below rather than being limited to the specifically described embodiments above.

What is claimed is:

1. A method of making a thermal interface material, comprising the steps of:
   (a) dispersing functionalized graphene sheets in a liquid;
   (b) removing the liquid through a filter so as to form a filtration cake of aligned functionalized graphene sheets, substantially all of which are parallel to a common plane;
   (c) cutting at least one block of aligned functionalized graphene sheets from the filtration cake, the block including a first end face and an oppositely-disposed second end face that are parallel to each other and to which substantially all of the functionalized graphene sheets are transverse, the block also including two oppositely-disposed sides to which substantially all of the functionalized graphene sheets are parallel; and
   (d) applying a first layer of a thermally conductive substance to the first end face and applying a second layer of the thermally conductive substance to the second end face of the block.

2. The method of claim 1, wherein the step of removing the liquid comprises the step of applying a filtration process to the liquid.

3. The method of claim 2, wherein the step of applying a filtration process comprises the steps of:
   (a) disposing the functionalized graphene sheets dispersed in the liquid on a first side filtration paper;
   (b) applying a vacuum to a second side of the filtration paper, wherein the second side is opposite from the first side, so as to draw the liquid through the filtration paper and away from the functionalized graphene sheets.

4. The method of claim 3, wherein the filtration paper comprises an aluminum oxide filtration paper.

5. The method of claim 1, wherein the liquid comprises water.

6. The method of claim 1, wherein the functionalized graphene sheets comprise functionalized multilayer graphene sheets.

7. The method of claim 1, wherein the functionalized graphene sheets comprise graphene functionalized with functional groups of a material selected from a group of materials consisting of: conjugated carbon-carbon, carbon-hydroxyls, carboxyls, and combinations thereof.

8. The method of claim 1, wherein the thermally conductive substance comprises indium.

9. The method of claim 8, further comprising the steps of:
   (a) applying a first silicon layer to the first layer of the thermally conductive substance; and
   (b) applying a second silicon layer to the second layer of the thermally conductive substance.
10. The method of claim 1, further comprising the steps of:
   (a) aligning a group of blocks of aligned functionalized
       graphene sheets so that the first end face of each block is
       substantially coplanar with the first end face of each other block;
   (b) applying a common first layer of a thermally conductive
       substance to the first end face of each block; and
   (c) applying a second common layer of the thermally con­
       ductive substance to the second end face of each block.

11. The method of claim 10, wherein the thermally con­
    ductive substance comprises indium.

12. The method of claim 10, further comprising the steps
    of:
   (a) applying a first silicon layer to the first layer of the
       thermally conductive substance; and
   (b) applying a second silicon layer to the second layer of
       the thermally conductive substance.

13. A method of making a thermal interface material for
    thermally coupling a heat source to a heat sink, comprising
    the steps of:
   (a) dispersing functionalized graphene sheets in a liquid;
   (b) removing the liquid through a filter so as to form a
       filtration cake of aligned functionalized graphene
       sheets, substantially all of which are parallel to a com­
       mon plane;
   (c) cutting at least one block of aligned functionalized
       graphene sheets from the filtration cake, the block
       including a first end face and an oppositely-disposed
       second end face that are parallel to each other and to
       which substantially all of the functionalized graphene
       sheets are transverse, the block also including two oppo­
       sitely-disposed sides to which substantially all of the
       functionalized graphene sheets are parallel;
   (d) applying a first layer of a thermally conductive sub­
       stance to the first end face and applying a second layer of
       the thermally conductive substance to the second end
       face of the block;
   (e) aligning a group of blocks of aligned functionalized
       graphene sheets so that the first end face of each block is
       substantially coplanar with the first end face of each other block;
   (f) applying a common first layer of a thermally conductive
       substance to the first end face of each block;
   (g) applying a second common layer of the thermally con­
       ductive substance to the second end face of each block;
   (h) applying a first silicon layer to the first layer of the
       thermally conductive substance; and
   (i) applying a second silicon layer to the second layer of the
       thermally conductive substance.

14. A method of making a thermal interface material, com­
    prising the steps of:
   (a) dispersing functionalized graphene sheets in a liquid;
   (b) removing the liquid through a filter so as to form a
       filtration cake of aligned functionalized graphene
       sheets, substantially all of which are parallel to a com­
       mon plane, wherein the removing of the liquid is per­
       formed through a filtration process that includes the
       steps of:
   (i) disposing the functionalized graphene sheets dis­
       persed in the liquid on a first side filtration paper; and
   (ii) applying a vacuum to a second side of the filtration
       paper, wherein the second side is opposite from the
       first side, so as to draw the liquid through the filtration
       paper and away from the functionalized graphene
       sheets;
   (c) cutting at least one block of aligned functionalized
       graphene sheets from the filtration cake, the block
       including a first end face and an oppositely-disposed
       second end face that are parallel to each other and to
       which substantially all of the functionalized graphene
       sheets are transverse, the block also including two oppo­
       sitely-disposed sides to which substantially all of the
       functionalized graphene sheets are parallel;
   (d) applying a first stabilizing layer to the first end face and
       a second stabilizing layer to the second end face; and
   (e) placing the first stabilizing layer to be in thermal com­
       munication with the heat source and the second stabiliz­
       ing layer to be in thermal communication with the heat
       sink.

15. The method of claim 14, wherein the filtration paper
    comprises an aluminum oxide filtration paper.

16. The method of claim 14, wherein the liquid comprises
    water.

17. The method of claim 14, wherein the functionalized
    graphene sheets comprise functionalized multilayer
    graphene sheets.

18. The method of claim 14, wherein the functionalized
    graphene sheets comprise graphene functionalized with func­
    tional groups of a material selected from a group of materials
    consisting of: conjugated carbon-carbon, carbon-hydroxyls,
    carboxyls, and combinations thereof.

19. The method of claim 14, wherein the thermally con­
    ductive substance comprises indium.

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