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(54) Title: METHODS OF FORMING CARBON-CARBON COMPOSITES USING ELECTRIC FIELD ASSISTED SINTERING AND RELATED CARBON-CARBON COMPOSITES

(57) Abstract: A method of forming a composite includes forming a composite preform through additive manufacturing. The method further includes pyrolyzing the composite preform to form a porous composite preform. The method also includes infiltrating the porous composite preform with a carbon precursor to form an infiltrated composite preform. The method further includes carbonizing the infiltrated composite preform to form a carbonized composite preform. The method also includes exposing the carbonized composite preform to electric field assisted sintering (EFAS).

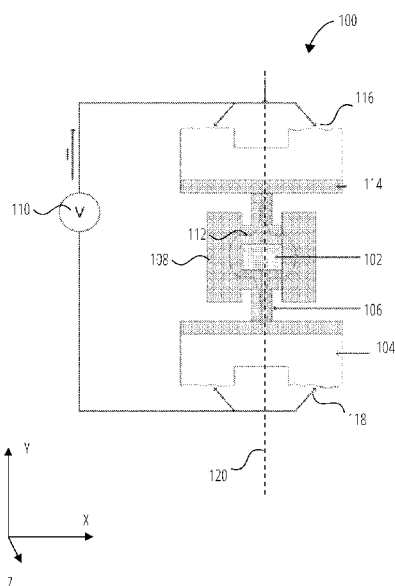


FIG. 1

SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

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**METHODS OF FORMING CARBON-CARBON COMPOSITES USING
ELECTRIC FIELD ASSISTED SINTERING AND RELATED CARBON-CARBON
COMPOSITES**

5 **CROSS-REFERENCE TO RELATED APPLICATION**

This application claims the benefit under 35 U.S.C. §119(e) of U.S. Provisional Patent Application Serial No. 63/378,775, filed October 7, 2022, the disclosure of which is hereby incorporated herein in its entirety by this reference.

10 **STATEMENT REGARDING FEDERALLY
SPONSORED RESEARCH OR DEVELOPMENT**

This invention was made with government support under Contract Number DE-AC07-05ID14517 awarded by the United States Department of Energy. The government has certain rights in the invention.

15 **TECHNICAL FIELD**

Methods of forming carbon-carbon composite materials are disclosed. More specifically, methods of forming carbon-carbon composite materials from additively manufactured carbon fiber preforms and using electric field assisted sintering are disclosed.

20 **BACKGROUND**

Manufacturing carbon-carbon (C-C) composite materials requires putting a carbon fiber preform through cycles of densification, carbonization, and graphitization processes. The cycles are then repeated until desired properties of the C-C composite material are achieved. Conventional graphitization processes are energy intensive as they involve heating a component to a temperature as high as 3000°C. In addition, the overall process of forming the C-C composite material is time consuming as heating rates are low and the heat may be limited by dimensions of the component since the heat is conducted throughout the component primarily by thermal diffusion. In addition to being energy intensive, C-C composite materials are expensive and time consuming to process.

Graphite tooling for electric field assisted sintering (EFAS) and spark plasma sintering (SPS) systems have mediocre mechanical properties, especially when sized up to make the tooling at industrially relevant scales. Graphite tooling die assemblies are made of high-density graphite and are considered a consumable component, often being replaced

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after a few uses. The temperatures and pressures used to achieve densification of the graphite use fine- or ultra-fine grained isostatically-molded graphite which is, by definition, isotropic in its mechanical and electrothermal properties. At small scale, the high purity isotropic graphite is not expensive and is easy to handle and machine. However, the cost of
5 high quality (e.g., high purity), graphite tooling at large scale is extremely expensive and has exceedingly long lead times. In addition, the use of graphite does not scale up in size well.

DISCLOSURE

10 Embodiments of the disclosure include a method of forming a composite. The method includes forming a composite preform through additive manufacturing. The method further includes pyrolyzing the composite preform to form a porous composite preform. The method also includes infiltrating the porous composite preform with a carbon precursor to form an infiltrated composite preform. The method further includes
15 carbonizing the infiltrated composite preform to form a carbonized composite preform. The method also includes exposing the carbonized composite preform to electric field assisted sintering (EFAS).

Another embodiment of the disclosure includes a composite structure. The composite structure includes carbon fibers embedded in a graphitized matrix. The
20 composite further includes a first region wherein the carbon fibers are oriented in a longitudinal orientation parallel to an axis of the composite structure. The composite also includes a second region wherein the carbon fibers are oriented in a lateral orientation perpendicular to the axis of the composite structure.

Other embodiments of the disclosure include a carbon-carbon composite. The
25 carbon-carbon composite includes a continuous carbon fiber embedded in a graphitized matrix, the carbon-carbon composite exhibiting a density of from about 1.60 g/cm³ to about 2.0 g/cm³.

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BRIEF DESCRIPTION OF THE DRAWINGS

While the specification concludes with claims particularly pointing out and distinctly claiming embodiments of the present disclosure, the advantages of embodiments of the disclosure may be more readily ascertained from the following description of
5 embodiments of the disclosure when read in conjunction with the accompanying drawings in which:

FIG. 1 illustrates a schematic representation of an electric field assisted sintering (EFAS) system in accordance with embodiments of the disclosure;

10 FIGS. 2 and 3 illustrate flow charts representative of methods of forming a composite structure in accordance with embodiments of the disclosure;

FIGS. 4A and 4B illustrate schematic representations of a fiber as part of an electrical system in accordance with embodiments of the disclosure;

FIG. 5 illustrates a composite structure in accordance with embodiments of the disclosure;

15 FIG. 6 illustrates a sectional view of a die assembly in accordance with embodiments of the disclosure; and

FIGS. 7A-7D illustrate examples of composite structures formed in accordance with embodiments of the disclosure.

20 MODE(S) FOR CARRYING OUT THE INVENTION

The following description provides specific details, such as material compositions, shapes, and sizes, in order to provide a thorough description of embodiments of the disclosure. However, a person of ordinary skill in the art would understand that the embodiments of the disclosure may be practiced without employing these specific details.
25 Indeed, the embodiments of the disclosure may be practiced in conjunction with conventional techniques employed in the industry.

Drawings presented herein are for illustrative purposes only, and are not meant to be actual views of any particular material, component, structure, device, or system. Variations from the shapes depicted in the drawings as a result, for example, of
30 manufacturing techniques and/or tolerances, are to be expected. Thus, embodiments described herein are not to be construed as being limited to the particular shapes or regions as illustrated, but include deviations in shapes that result, for example, from manufacturing. For example, a region illustrated or described as box-shaped may have rough and/or

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nonlinear features, and a region illustrated or described as round may include some rough and/or linear features. Moreover, sharp angles that are illustrated may be rounded, and vice versa. Thus, the regions illustrated in the figures are schematic in nature, and their shapes are not intended to illustrate the precise shape of a region and do not limit the scope of the present claims. The drawings are not necessarily to scale. Additionally, elements common
5 between figures may retain the same numerical designation.

As used herein, the terms “configured” and “configuration” refers to a size, a shape, a material composition, a material distribution, orientation, and arrangement of at least one feature (e.g., one or more of at least one structure, at least one material, at least one region,
10 at least one device) facilitating use of the at least one feature in a pre-determined way.

As used herein, the term “substantially” in reference to a given parameter means and includes to a degree that one skilled in the art would understand that the given parameter, property, or condition is met with a small degree of variance, such as within acceptable manufacturing tolerances. By way of example, depending on the particular
15 parameter, property, or condition that is substantially met, the parameter, property, or condition may be at least 90.0 percent met, at least 95.0 percent met, at least 99.0 percent met, at least 99.9 percent met, or even 100.0 percent met.

As used herein, “about” or “approximately” in reference to a numerical value for a particular parameter is inclusive of the numerical value and a degree of variance from the numerical value that one of ordinary skill in the art would understand is within acceptable tolerances for the particular parameter. For example, “about” or “approximately” in
20 reference to a numerical value may include additional numerical values within a range of from 90.0 percent to 110.0 percent of the numerical value, such as within a range of from 95.0 percent to 105.0 percent of the numerical value, within a range of from 97.5 percent to 102.5 percent of the numerical value, within a range of from 99.0 percent
25 to 101.0 percent of the numerical value, within a range of from 99.5 percent to 100.5 percent of the numerical value, or within a range of from 99.9 percent to 100.1 percent of the numerical value.

As used herein, relational terms, such as “beneath,” “below,” “lower,” “bottom,” “above,” “upper,” “top,” “front,” “rear,” “left,” “right,” and the like, may be used for ease
30 of description to describe one element’s or feature’s relationship to another element(s) or feature(s) as illustrated in the drawings. Unless otherwise specified, the spatially relative terms are intended to encompass different orientations of the materials in addition to the

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orientation depicted in the figures. For example, if materials in the figures are inverted, elements described as “below” or “beneath” or “under” or “on bottom of” other elements or features would then be oriented “above” or “on top of” the other elements or features. Thus, the term “below” can encompass both an orientation of above and below, depending
5 on the context in which the term is used, which will be evident to one of ordinary skill in the art. The materials may be otherwise oriented (e.g., rotated 90 degrees, inverted, flipped) and the spatially relative descriptors used herein interpreted accordingly.

As used herein, the singular forms “a,” “an,” and “the” are intended to include the plural forms as well, unless the context clearly indicates otherwise.

10 As used herein, the term “and/or” means and includes any and all combinations of one or more of the associated listed items.

As used herein, the terms “vertical,” “longitudinal,” “horizontal,” and “lateral” are in reference to a major plane of a structure and are not necessarily defined by earth’s gravitational field. A “horizontal” or “lateral” direction is a direction that is substantially
15 parallel to the major plane of the structure, while a “vertical” or “longitudinal” direction is a direction that is substantially perpendicular to the major plane of the structure. The major plane of the structure is defined by a surface of the structure having a relatively large area compared to other surfaces of the structure. With reference to the drawings, a “horizontal” or “lateral” direction may be perpendicular to an indicated “Z” axis, and may be parallel to
20 an indicated “X” axis and/or parallel to an indicated “Y” axis; and a “vertical” or “longitudinal” direction may be parallel to an indicated “Z” axis, may be perpendicular to an indicated “X” axis, and may be perpendicular to an indicated “Y” axis.

As used herein, the terms “electrical field assisted sintering (EFAS)” and “spark plasma sintering (SPS)” are equivalent terms that may be used interchangeably.

25 A carbon-carbon (C-C) composite is formed from a carbon fiber preform that is formed by additive manufacturing and subjected to electric field assisted sintering (EFAS). The carbon fiber preform is pyrolyzed to form a porous carbon fiber preform, which is infiltrated with a carbon precursor to form an infiltrated carbon fiber preform. The infiltrated carbon fiber preform is carbonized to form a carbonized carbon fiber preform,
30 which is graphitized in an EFAS system to form the C-C composite. The EFAS system may function as a source of heat and energy for the graphitization. Using the EFAS process in the graphitization act also densifies the carbonized carbon fiber preform. C-C composites may be densified using EFAS faster than by conventional processes.

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The graphitization process conducted on the carbon fiber preform provides a more ready conversion to graphite from carbon than conventional graphitization processes. In addition, the graphitized carbon fiber preform is formed without using a catalyst. A density of the resulting C-C composite is tailorable by adjusting process conditions used during the formation of the C-C composite, such as by adjusting pressure conditions and/or temperature conditions during graphitization by EFAS. The process of forming the C-C composite is also less costly than conventional techniques for forming C-C composites and replaces the need to design and fabricate an ultra-high temperature furnace system.

FIG. 1 illustrates a schematic view of an EFAS system 100 configured to form a part 102. The EFAS system 100 includes one or more rams 104 configured to apply pressure to the part 102 in a direction along an axis 120 of the EFAS system 100 (e.g., in a direction along the Y axis) through one or more plungers 106. A die 108 may be configured to define a laterally outer shape of the part 102 (e.g., in the X and Z directions), while the plungers 106 may be configured to define vertically outer shapes of the part 102 (e.g., in the Y direction). While the embodiment illustrated in FIG. 1 illustrates two opposing rams 104, other embodiments may include a single ram 104 on one side of the EFAS system 100 with a non-moving structure, such as an anvil positioned opposite the ram 104.

The EFAS system 100 includes a voltage source 110, such as line power, a power inverter, a battery, a generator, etc., configured to induce a current through the EFAS system 100. The voltage source 110 is coupled to a first electrode 116 and a second electrode 118 on opposing sides of the EFAS system 100, such that a voltage applied between the first electrode 116 and the second electrode 118 generate a current from a first ram 104 to a second ram 104 across the plungers 106, die 108, and the part 102. The current generated by the voltage source 110 may follow current paths 112 through the plungers 106 and the die 108. The plungers 106 and the die 108 may be formed from a material configured to generate heat through resistance to the passage of the current through the respective plunger 106 or die 108. Some of the current may also pass across the part 102. The part 102 may also generate heat through resistance to the current passing therethrough. The heat generated by the current passing through the part 102, the plungers 106, and the die 108 may heat the part 102 as part of the sintering process.

The plungers 106 and dies 108 may be referred to generally herein as tooling. The tooling may be formed from composite materials. The formation processes for the tooling

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may facilitate controlling the current paths 112 through the tooling, such that the material of the tooling is configured to concentrate the generated heat in a region near the part 102. Furthermore, controlling the current paths 112 may facilitate increasing a size of the plungers 106 and die 108 at least by facilitating improved control over the distribution of
5 heat within the tooling. Increasing a size of the plungers 106 and die 108 may similarly facilitate using the EFAS system 100 to form larger parts 102.

The rams 104 may include spacers 114 positioned between the rams 104 and the plungers 106. The spacers 114 may be configured to thermally insulate the rams 104 from the heat generated in the plungers 106. For example, the spacers 114 may be formed from
10 an electrically conductive material having high hardness that is also thermally insulative, such as graphite or a carbon-carbon composite.

The EFAS system 100 may be configured to provide pulsed or continuous DC power through the voltage source 110. The EFAS system 100 may be commercially available, such as a DCS-5 system available from Thermal Technology, LLC (Santa Rosa,
15 CA). Alternatively, if the part 102 to be formed exhibits large dimensions, such as having a diameter of up to about 0.6 m, a system with a pressing load of up to 800 metric tons and an applied current of 150,000A may be used to conduct the graphitization process.

FIGS. 2 and 3 illustrate flow charts representative of methods 200, 300 of forming C-C composites, such as the plungers 106 and dies 108 described above. As discussed
20 above, the tooling may be formed from composite materials. FIG. 2 describes a method 200 of forming C-C composites using a preform composite material, such as a carbon fiber preform.

A composite preform is subjected to a pyrolyzation act in act 202, one or more infiltration acts 204, one or more carbonization acts 206, and a graphitization act 208 to
25 form the composite, such as a carbon-carbon composite (e.g., a carbon fiber carbon composite) or a carbon-silicon carbide (C-SiC) composite. While several embodiments herein describe a carbon-carbon composite and forming a carbon-carbon composite, a similar process may be used to form other carbon composites, such as carbon-silicon carbide (C-SiC) composites or carbon-boron carbide (C-BC) composites.

30 The composite preform may include fibers (such as carbon fibers, silicon fibers, etc.) and a resin binding the fibers into the preform shape. The fibers include continuous fibers that are embedded within the resin. The resin may include, but is not limited to, a thermoset polymer, a single- or a multi-part epoxy resin, a polyester resin, a cationic epoxy,

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an acrylated epoxy, a urethane, an ester, a thermoplastic polymer, a photopolymer, a polyepoxide, a thiol, an alkene, or a thiol-ene. The resin may be formulated to be fully convertible to carbon during pyrolysis without significant shape change occurring to the fiber preform. By way of example only, the resin may be a UV curable resin. The
5 composite preform may be selected from a commercially available composite preform, which are available from numerous sources, such as from Continuous Composites (Coeur d'Alene, ID).

During the pyrolyzation act in act 202, the composite preform is pyrolyzed to remove the resin, resulting in a porous composite preform. The porous composite preform
10 includes a network of pores throughout the composite preform. The pyrolyzation may be conducted at a temperature sufficient to pyrolyze the resin, which temperature depends on the composite preform and resin used. Removing the resin leaves behind carbon compounds and forms the pores in the composite preform, which may be of sufficient size and sufficient distribution to retain a carbon precursor during the infiltration act. The
15 composite preform may be pyrolyzed in a conventional furnace, such as in a box furnace under a nitrogen environment.

The carbon precursor may be introduced (e.g., infiltrated, impregnated) into the pores of the porous composite preform to form an infiltrated composite preform in act 204. The porous composite preform may be infiltrated with the carbon precursor in a
20 conventional furnace, such as in a box furnace. The carbon precursor is carbon-rich and is formulated to incorporate additional carbon into the porous composite preform relative to an initial carbon content in the composite preform. The carbon precursor may be a carbon yielding precursor that is formulated to increase the carbon content in the infiltrated composite preform after conducting the one or more infiltration processes. While FIG. 2
25 illustrates a single infiltration process, multiple infiltration processes may be conducted. The carbon precursor may exhibit a low viscosity, enabling easy infiltration into the pyrolyzed composite preform. The carbon precursor may include, for example, pitch, such as a mesophase pitch. In some embodiments, the carbon precursor is a naphthalene-based mesophase pitch. However, other carbon precursors may be used. The carbon precursor
30 may be impregnated into the pores by conventional techniques, such as by vacuum infiltration. However, other infiltration techniques may be used. In some embodiments, the porous composite preform is infiltrated with the naphthalene-based mesophase pitch. In some embodiments, where a carbon composite is being formed, a carbon fiber preform

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is used and the carbon precursor is infiltrated into pores formed in the carbon fiber preform after the pyrolyzation act 202. In other embodiments, such as where a C-SiC composite is to be formed, a silicon carbide precursor may be infiltrated into the pores of the porous carbon fiber preform.

5 The infiltrated composite preform may be carbonized, in act 206, to form a carbonized composite preform that includes additional carbon relative to the initial carbon content of the composite preform. The infiltrated composite preform may be subjected to heat (e.g., a thermal process) in argon (e.g., an inert gas) to remove resin organics and increase the carbon content. Carbon species of the carbon precursor may, therefore, remain
10 (e.g., be retained) in the carbonized composite preform following the carbonization process. In addition to increasing the carbon loading, the density of the infiltrated composite preform may be reduced due to outgassing and carbon foam extrusion. The infiltrated composite preform may be carbonized in a conventional furnace, such as in a box furnace under a nitrogen environment.

15 Multiple infiltration acts and carbonization acts may be conducted to increase the carbon loading of the infiltrated composite preform to a desired carbon loading. The infiltration acts and carbonization acts may be conducted in conventional furnaces used in C-C composite manufacturing. Temperature and time conditions for the infiltration acts and the carbonization acts may be determined based on the desired density of the C-C
20 composite to be formed.

 After a desired degree of infiltration and carbon loading are achieved, the carbonized composite preform is graphitized by EFAS to form the C-C composite in act 208. The EFAS process graphitizes the carbonized composite preform, with the EFAS system functioning as the source of heat (e.g., energy) for the graphitization. The
25 carbonized composite preform is graphitized to produce a densified, composite component having a graphitized matrix. The EFAS system 100 (FIG. 1) may be configured to apply a high electrical current that quickly heats the carbonized composite preform under the simultaneous application of a uniaxial pressure inside of a vacuum chamber. A schematic of an EFAS system 100 according to embodiments of the disclosure is described above,
30 and illustrated in FIG. 1, with the indicated part 102 (FIG. 1) corresponding to the carbonized composite preform before the graphitization process is conducted.

 An electric current (e.g., current density) is applied to the carbonized composite preform, which is positioned between an upper plunger 106 (FIG. 1) and a lower

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plunger 106 (FIG. 1) of the EFAS system 100 (FIG. 1). A magnitude of the electric current used depends on a desired final temperature of the carbonized composite preform, as well as on material properties, die geometry, geometry of the upper plunger 106, geometry of the lower plunger 106, and the size of the carbonized composite preform. A first
5 electrode 116 (FIG. 1) and a second electrode 118 (FIG. 1) of the EFAS system 100 are used to apply the electric current. The electric current flowing through the plungers 106 (FIG. 1) (e.g., upper plunger 106, lower plunger 106) may range from about 1240 amps (A) to about 150,000A, such as from about 1240A to about 140,000A, from about 1300A to about 130,000A, or from about 1325A to about 130,000A. Heat and pressure are applied
10 to the carbonized composite preform during the graphitization process of act 208, with the applied force being in an axial direction along the Y axis. Joule heating of the carbonized composite preform occurs, facilitating high heating rates and causing the carbonized composite preform to be heated from the inside. The joule heating facilitates the carbonized composite preform to be heated to a temperature of greater than about 2000°C,
15 while other portions of the EFAS system 100, such as the rams 104 and electrodes 116, 118, remain at a lower temperature, such as less than about 400°C. The joule heating is facilitated by conductivity (e.g., thermal and electrical conductivity) of the carbonized composite preform.

The temperature and pressure conditions used in the graphitization process may be
20 selected to graphitize and densify the carbonized composite preform. The temperature to which the carbonized composite preform is exposed may range from about 1000°C to about 3000°C, such as from about 1500°C to about 3000°C, from about 1800°C to about 2800°C, from about 2000°C to about 2500°C, or from about 2000°C to about 3000°C. The temperature may start at an initial temperature and be subsequently
25 increased. By way of example only, the carbonized composite preform may be heated at a heating profile of about 100°C/min up to about 1500°C, then at a heating profile of about 50°C/min to about 2000°C, and then at a heating profile of about 33°C/min to a final graphitization temperature of about 2500°C. The carbonized composite preform may be maintained at an internal temperature of about 2500°C for a sufficient amount of time to
30 graphitize the carbon precursor. The overall time of the graphitization process may be from about 0.5 hour to about 5 hours, while the final graphitization temperature may be maintained at from about 5 minutes to about 30 minutes. The pressure applied during the graphitization process may be from about 5 MPa to about 500 MPa, such as from about 5

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MPa to about 300 MPa, from about 5 MPa to about 100 MPa, from about 5 MPa to about 50 MPa, from about 5 MPa to about 30 MPa, from about 10 MPa to about 100 MPa, from about 15 MPa to about 100 MPa, from about 20 MPa to about 50 MPa, from about 30 MPa to about 50 MPa, or from about 40 MPa to about 80 MPa. The pressure used in the
5 graphitization process may be sufficient to fully densify the carbonized carbon fiber preform to form the C-C composite. As the pressure is applied, channels and voids within the C-C composite may be collapsed and fused.

Since the carbonized composite preform is heated from its interior, an amount of time used to graphitize the carbonized carbon fiber preform is significantly less than an
10 amount of time used to conduct graphitization in a conventional furnace, which requires heating of the preform and its surrounding environment and subsequent diffusion of the heat into the preform. The amount of time to graphitize a carbonized carbon fiber preform using the EFAS process according to embodiments of the disclosure may be about one-half of the amount of time used to graphitize a similar carbonized carbon fiber preform using a
15 conventional furnace. Therefore, using the EFAS process to graphitize the carbonized composite preform eliminates the thermal diffusion time constraint associated with conventional techniques.

An amount of energy used to graphitize the carbonized composite preform in the EFAS process according to embodiments of the disclosure is also significantly less than an
20 amount of energy used to conduct graphitization in a conventional furnace. Therefore, the graphitization process according to embodiments of the disclosure is more efficient than conventional graphitization techniques using a furnace. The graphitization process using EFAS may achieve a reduction in energy consumption from about 50% to about 85% relative to that of a furnace.

25 Conventional graphitization processes do not increase the density of its resulting articles. However, the density of the C-C composite formed according to embodiments of the disclosure is increased simultaneously during the graphitization process using EFAS at least partially due to the pressure applied by the EFAS system 100 (FIG. 1) compressing the carbonized composite preform along the axis 120 (FIG. 1) of the EFAS system 100
30 (FIG. 1). Internal porosity within the graphitized composite preform collapses and becomes fused during the graphitization process. By increasing the pressure applied during the graphitization process, the porosity within the graphitized composite preform is decreased and the density is increased. In some cases, adjacent fibers in the composite

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preform fuse together due at least in part to high localized current densities and the associated Joule heating at fiber contacts. The fused fibers create a bonded multi-directional network of reinforced fibers further increasing a density of the composite preform. The uniaxial pressure applied during the graphitization process may, therefore, be
5 tailored to achieve a desired compression of the carbonized composite preform following the graphitization process.

The graphitization process according to embodiments of the disclosure may be conducted in less than or equal to about 10 hours, such as less than or equal to about 5 hours, less than or equal to about 2 hours, or less than or equal to about 1 hour. In contrast,
10 conventional graphitization techniques, such as in a furnace, may take up to multiple weeks. The reduction in time may facilitate the formation of composites having higher degrees of graphitization, such in a range from about 50% graphitization to about 90% graphitization, or from about 50% graphitization to about 80% graphitization.

The resulting C-C composite may exhibit a density of up to about 2 g/cm^3 , such as
15 from about 1.50 g/cm^3 to about 2 g/cm^3 or from about 1.60 g/cm^3 to about 1.92 g/cm^3 . The density of the C-C composite may be relatively greater than the density of the associated composite preform. While the density of the composite preform may initially decrease during the pyrolyzation act, the density may increase during the one or more infiltration acts. The density may decrease by a small amount during the one or more carbonization
20 acts, as some of the carbon precursor may be exuded due to outgassing. The density of the C-C composite may, however, achieve its final density, which is greater than the density of the composite preform, after conducting the one or more infiltration acts, the one or more carbonization acts, and the graphitization act.

The overall formation of the C-C composite according to embodiments of the
25 disclosure may be completed in a shorter amount of time than conventional processes, which take months to produce a C-C composite of a similar density. By way of example only, a C-C composite having a density of 1.50 g/cm^3 may be formed according to embodiments of the disclosure within about one week. A first pyrolyzation act may be conducted in about 1 day, a first infiltration act may be conducted in about 1 day, a first
30 carbonization act may be conducted in about 1 day, a second infiltration act may be conducted in about 1 day, a second carbonization act may be conducted in about 1 day, and a graphitization act may be conducted in about 0.5 day. In contrast, conventional processes of forming a C-C composite having a similar density take months, such as about 2 months,

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to complete. Furthermore, the C-C composite may be formed by conducting significantly fewer cycles of infiltration and carbonization and a single graphitization cycle, compared to conventional processes in which significantly more cycles are conducted. By way of example only, a C-C composite according to embodiments of the disclosure exhibiting a
5 density of greater than about 1.6 g/cm^3 was achieved after conducting only two cycles of infiltration and a single graphitization act.

The degree of graphitization and the density of the resulting C-C composite may be controlled by controlling individual parameters during the graphitization process, such as the pressure applied, the temperature of the part, and the time that the part is maintained
10 and the desired temperature and pressure.

FIG. 3 illustrates a flow chart representative of a method 300 of forming C-C composites including manufacturing the composite preform in act 302. The composite preform may be formed in an additive manufacturing process (e.g., 3-D printing process). Similar to the process described above, with respect to FIG. 2, the composite preform is
15 pyrolyzed in act 304, infiltrated with a carbon precursor in act 306, carbonized in act 308 and graphitized in act 310.

Forming the composite preform by additive manufacturing may facilitate the carbon precursor infiltrated into the pyrolyzed composite preform in act 306 to be better retained during the carbonization act 308. The additively manufactured composite preform may be
20 highly ordered and densely packed, which improves retention of the carbon precursor during carbonization. The additive manufacturing of the composite preform also enables near-net shapes of the carbon fiber preform to be printed, which reduces post-processing time and machining time to produce the C-C composite at a desired shape or geometry. The additive manufacturing of the carbon fiber preform may also facilitate controlled
25 shapes (e.g., geometries) and sizes of the C-C composite to be formed. Therefore, custom C-C composites, such as die assemblies, may be formed. The die assemblies may include gas flow ports and passages to condense and drain liquid-rich effluent during the manufacture of the C-C composite.

The additive manufacturing of the composite preform also provides enhanced
30 electro-thermal control of process parameters. For example, additive manufacture may facilitate improved control over fiber orientation within the composite preform. As discussed in further detail below, different fiber orientations may change thermal properties of the associated part and electrical conductivity properties of the associated part in

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different regions of the part. For example, a first fiber orientation may exhibit a high thermal conductivity and low electrical resistance. The high thermal conductivity may cause heat generated in the part or near the part to distribute quickly and evenly through the part. The low electrical resistance may cause relatively little heat to be generated as the
5 electrical current passes across the part. Another orientation may exhibit low thermal conductivity and high electrical resistance. The low thermal conductivity may cause the material to have high insulative properties (e.g., a high resistance to heat transfer through the part). The high electrical resistance may cause relatively high amounts of heat to be generated when the electrical current passes through the part. Controlling the flow of heat
10 and electrical current within the composite preform may facilitate improved control of the heating of the composite preform during the graphitization process of act 310. The fiber orientation may also facilitate the control of the flow of heat and electrical current in a final C-C composite part, which may be used as tooling in an EFAS system 100 (FIG. 1).

The C-C composite according to embodiments of the disclosure may, for example,
15 be used as tooling in CMC/CFC manufacturing as a replacement for conventional 100% graphite tooling, which exhibits non-ideal properties. For example, the C-C composite may be configured as a carbon-carbon (C-C) die and plunger assembly as a replacement for conventional 100% graphite die and plunger assemblies. The C-C composite may be used as tooling in EFAS systems. By replacing the conventional graphitic die and plunger
20 assemblies with the C-C composite, the use of non-graphitic tooling (e.g., composite tooling) will enable greater achievable loads, heating rates (through lessened thermal gradients), and uniformity of samples at up to about 0.2 m diameter. In addition, the plungers of the die assemblies may be manufactured with desired fiber orientations, such as a quasi-isotropic (QI) orientation, a unidirectional orientation, or a spiral wound (SW)
25 orientation. The fiber orientation may be used to achieve desired properties of the C-C composite along the respective orientation.

The graphitization process described above may also be used to repair a damaged C-C composite. For example, if the C-C composite has delaminated, become bloated, or otherwise damaged during processing, during transport, or during storage, the damaged C-
30 C composite may be placed in the EFAS system and the graphitization process conducted to repair the defects. Heat and pressure produced by the EFAS system may be applied to the damaged C-C composite as described above. The graphitization process may be used to repair a damaged C-C composite formed by the process described above or formed by

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another (e.g., a conventional) process. In other words, the graphitization process may be used to repair a C-C composite formed using EFAS or to repair a conventional C-C composite.

After conducting the graphitization process, the damaged C-C composite may exhibit a density of up to about 2 g/cm³. The repair process according to embodiments of the disclosure may be conducted in less than a day, compared to weeks for conventional repair techniques or having to discard the damaged C-C composite.

To further increase carbon loading of the C-C composite, carbon black or graphene may be used with the mesophase pitch to infiltrate (e.g., impregnate) the pyrolyzed composite preform. The carbon black or graphene may enable higher carbon conversions to be achieved during the carbonization act. The carbon black or graphene may be combined with the mesophase pitch or other carbon precursor and infiltrated into the pyrolyzed composite preform as described above. Alternatively, the mesophase pitch or other carbon precursor and the carbon black or graphene may be separately infiltrated into the pyrolyzed composite preform. The carbon black or graphene may also function as a filler. Other carbon-containing fillers may also be used instead of, or in addition to, carbon black or graphene. By incorporating carbon black, graphene, or another filler with the carbon precursor, a low viscosity material may be infiltrated into the pyrolyzed composite preform. The carbon conversion efficiency of the C-C composite is increased because the direct rule of mixtures increases carbon content due to the presence of the carbon black or graphene. The carbon black or graphene also provides well-dispersed and numerous nucleation sites.

The graphitization processes described above may be used to produce larger scale C-C composites than those produced by conventional techniques, which produce components at a small scale, such as having a diameter of less than about 20 mm. For instance, the C-C composite according to embodiments of the disclosure may have a diameter of from about 0.1 m (about 100 mm) to less than or equal to about 1.0 m (about 1000 mm), such as from about 0.1 m to less than or equal to about 0.6 m (about 600 mm), from about 0.1 m to less than or equal to about 0.5 m (about 500 mm), or from about 0.1 m to less than or equal to about 0.2 m (about 200 mm). The ability to scale up to larger dimensions (e.g., a large diameter) was surprising, at least because conventional EFAS techniques and EFAS systems have limitations relating to tooling (die and plunger) size, strength of the tooling, and the presence of thermal and other process gradients. The

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C-C composite according to embodiments of the disclosure may also exhibit a greater uniformity of composition at the larger scale than may be produced by conventional EFAS techniques. The C-C composite may exhibit improved mechanical properties, such as strength (burst, crush), hardness, and electrothermal properties, even at the larger scale, compared to commercially available grades of graphite, such as Mersen 2160, Tokai G535, etc. Production rates of the larger diameter C-C composites may be higher than those of smaller diameter C-C composites produced by conventional EFAS techniques and EFAS systems.

The C-C composites produced according to embodiments of the disclosure may be less than half the cost of conventional C-C composites, and may exhibit superior strength, thermal conductivity, and fracture toughness compared to conventional C-C composites. The C-C composites may be used as advanced and/or refractory materials. The improved properties greatly enhance the operational breadth of conventional C-C composites for aerospace and defense applications, as well as other applications. By way of example only, the C-C composites may be used as brake discs, rocket nozzles, nose cones, or leading edges of wings. The C-C composites may also be used as tooling in the EFAS system instead of conventional graphite tooling. The C-C composites may also be used in energy storage and production, biomaterials, advanced composites, optical materials, ultra-high temperature ceramics and metals, tough ceramics, high strength and high melting temperature (T_m) metals, ultra-high temperature heat exchangers, solid oxide fuel cell/solid oxide electrolytic cells assemblies, and multi-part sub-assemblies.

As discussed above, an orientation of fibers, such as carbon fibers or graphite fibers, within a composite may affect the electrical and thermal properties of the associated composite. FIGS. 4A and 4B illustrate schematic views of a single fiber 402a, 402b in different orientations. A voltage source 404 is configured to apply a voltage across the associated fiber 402a, 402b and generate a current across the associated fiber 402a, 402b along the current path 410.

FIG. 4A illustrates the voltage source 404 configured to apply a current through the fiber 402a longitudinally (e.g., along a longitudinal axis) between a first axial end 406 to a second axial end 408. The fiber 402a may be configured to have high electrical conductivity in a longitudinal direction. Thus, the current flowing through the fiber 402a between the first axial end 406 and the second axial end 408 may meet with little resistance. The low resistance in the fiber 402a may result in little to no heat generation in

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the fiber 402a from the flow of electricity through the fiber 402a. The heat generated in the fiber 402a may correlate to the power used to create the current through the fiber 402a, such that the power to transmit the current through the fiber 402a is also relatively low.

FIG. 4B illustrates the voltage source 404 configured to apply the current through
5 the fiber 402b laterally (e.g., perpendicular to the longitudinal axis) between a first lateral edge 412 and a second lateral edge 414. The fiber 402b may be configured to have a high electrical resistance in the lateral direction. Thus, the current flowing through the fiber 402b between the first lateral edge 412 and the second lateral edge 414 may meet with a large amount of resistance. The high resistance may generate large amounts of heat as
10 the current passes laterally through the fiber 402b. A relatively higher amount of power may also be used to generate the current and overcome the resistance in the lateral direction.

The fibers 402a, 402b may also exhibit lower resistance to heat transfer in a longitudinal direction than in the lateral direction. Therefore, similar to electricity, the heat
15 may transfer with relative ease through the associated fiber 402a, 402b in the longitudinal direction while the heat may be met with significantly more resistance in the lateral direction. Therefore, heat may be substantially evenly distributed across a longitudinal length of the fibers 402a, 402b (e.g., between the first axial end 406 and the second axial end 408), while the heat may have a higher gradient laterally across the fibers 402a, 402b
20 (e.g., between the first lateral edge 412 and the second lateral edge 414).

Selectively positioning (e.g., orienting) the fibers 402a, 402b within a part may facilitate better control over where heat is generated in a part when a current is applied thereto. Selectively positioning the fibers 402a, 402b may also reduce the power consumed by the parts during an EFAS process, such as by reducing power consumption in areas of a
25 part or tool where little to no heat generation is needed and by insulating the regions where heat generation is needed to reduce heat losses.

FIG. 5 illustrates a composite structure 500 formed from a composite material including selectively positioned fibers 502a, 502b. The composite structure 500 may be configured to have a voltage from a voltage source 504 applied from a first axial end 506 to a second axial end 508, such that a current flows across the composite structure 500 from
30 the first axial end 506 to the second axial end 508.

In the embodiment illustrated in FIG. 5, the fibers 502a in a first portion of the composite structure 500 are positioned in a longitudinal orientation, such that the current

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flows longitudinally along a longitudinal axis of the individual fibers 502a. As discussed above, the longitudinal orientation of the fibers 502a results in a relatively low resistance to the current. Therefore, the fibers 502a in the first portion of the composite structure 500 combine to form a low resistance region 510 of the composite structure 500.

5 The fibers 502b in a second portion of the composite structure 500 are positioned in a lateral orientation. As illustrated in FIG. 5, the fibers 502b in the second portion are wound about a longitudinal axis of the composite structure 500, such that current flowing axially through the composite structure 500 passes laterally through the fibers 502b. As discussed above, the lateral orientation of the fibers 502b results in a relatively high
10 resistance to the current passing therethrough. Therefore, the fibers 502b in the second portion of the composite structure 500 combine to form a high resistance region 512 of the composite structure 500.

 The high resistance region 512 is configured to generate large amounts of heat due to the current passing through the high resistance of the laterally oriented fibers 502b in the
15 high resistance region 512. The low resistance region 510 is configured to separate the high resistance region 512 from other components of the system (e.g., an EFAS system), while providing a much lower resistance to the flow of current, which may reduce power consumption through the composite structure 500 and reduce the heat of the first axial end 506.

20 The composite structure 500 may be configured to act as a part of an EFAS system, such as a plunger of the EFAS system. For example, the high resistance region 512 may be the portion configured to contact a part being formed by the EFAS system within an associated die. The high resistance region 512 is configured to generate heat in a region proximate the part being formed. The low resistance region 510 may be configured to
25 separate the rams of the EFAS system from the high temperatures in the high resistance region 512, such that the rams may be formed from different materials with a lower heat tolerance, which may be less expensive.

 FIG. 6 illustrates a sectional view of an assembly 600 for an EFAS system (e.g., EFAS system 100) formed from composite structures, such as composite structure 500.
30 The assembly 600 includes a die 602 defining a cavity 612 within which a part 606 is positioned to be formed through an EFAS process. Two plungers 604 are positioned on opposing axial sides of the cavity 612 and are configured to extend into the cavity 612 and

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apply pressure on upper and lower surfaces of the part 606 within the cavity 612 in a direction parallel to an axis 614 of the assembly 600.

The plungers 604 may be formed in a similar manner to the composite structure 500 of FIG. 5. For example, the plungers 604 may include a high resistance region 608 and a low resistance region 610. The plungers 604 are arranged to position the high resistance region 608 adjacent to the part 606 in the cavity 612. Positioning the high resistance region 608 adjacent to the part 606 may concentrate the heat generated in the plunger 604 by the current flowing therethrough in a region near the part 606. Concentrating the heat in the region near the part may raise a temperature of the part 606 to the desired temperature quickly and may result in a reduction of power used to generate the heat by not generating heat in the low resistance region 610 of the plungers 604.

The fibers in the die 602 may be arranged in a similar manner to the high resistance region 608 of the plungers 604, such that the die 602 is configured to be a high resistance structure in the axial direction. Configuring the die 602 to be a high resistance structure in the axial direction may result in the die 602 generating large amounts of heat, similar to the high resistance region 608 of the plungers 604, when a current is applied axially to the assembly 600. In other embodiments, the arrangement of the fibers in the die 602 may be different at different distances from the axis 614 of the assembly 600. For example, radially inward portions of the die 602 may have fibers positioned in a lateral orientation and radially outward portions of the die 602 may have fibers positioned in a longitudinal orientation. The laterally oriented fibers in the radially inward portions of the die 602 may be configured to generate heat in the region adjacent to the cavity 612 where the part 606 is being formed and the longitudinally oriented fibers in the radially outward portions of the die 602 may be configured to resist the transmission of heat from the radially inward portions of the die 602 to maintain the heat generated in the radially inward portions of the die 602 in the region adjacent to the cavity 612.

FIGS. 7A-7D illustrate examples of composite structures 702, 708, 714, 720 formed in accordance with embodiments of the disclosure. The composite structures 702, 708, 714, 720 illustrated in FIGS. 7A-7D illustrate a variety of shapes and orientations. However, the shapes and orientations illustrated in FIGS. 7A-7D are exemplary only and in no way limiting. It is noted that many different and more complex shapes and orientations may be formed through the same processes described herein.

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FIG. 7A and FIG. 7B illustrate composite structures 702, 708 having a cylindrical shape. In the composite structure 702 of FIG. 7A, the fibers 704 are arranged in a lateral arrangement, such that the fibers are arranged perpendicular to a longitudinal axis 706 of the composite structure 702. As discussed above, an electrical current passing across the
5 composite structure 702 along the longitudinal axis 706 would encounter a high resistance due to the lateral arrangement of the fibers 704. Thus, the composite structure 702 of FIG. 7A would generate a large amount of heat when an electrical current passes across the composite structure 702 along the longitudinal axis 706.

In the composite structure 708 of FIG. 7B, the fibers 710 are arranged in a
10 longitudinal arrangement, such that the fibers are arranged parallel to a longitudinal axis 712 of the composite structure 708. As discussed above, an electrical current passing across the composite structure 708 along the longitudinal axis 712 would encounter a low resistance due to the longitudinal arrangement of the fibers 710. Thus, the composite structure 708 of FIG. 7B would generate little heat when an electrical current passes across
15 the composite structure 708 along the longitudinal axis 712. As discussed above, a single cylindrical composite structure may incorporate more than one orientation of fibers, such as the composite structure 500 (FIG. 5), which includes a low resistance region 510 with fibers arranged longitudinally similar to the composite structure 708 and a high resistance region 512 with fibers arranged laterally similar to the composite structure 702.

20 FIG. 7C illustrates composite structure 714 having a rectangular cylinder shape. The fibers 716 are arranged in a lateral arrangement, such that the fibers are arranged perpendicular to a longitudinal axis 718 of the composite structure 714. As discussed above, an electrical current passing across the composite structure 714 along the longitudinal axis 718 would encounter a high resistance due to the lateral arrangement of
25 the fibers 716. Thus, the composite structure 714 of FIG. 7C would generate a large amount of heat when an electrical current passes across the composite structure 714 along the longitudinal axis 718.

FIG. 7D illustrates a composite structure 720 having a more complex “hour glass” shape having laterally wider portions on opposing longitudinal ends of the composite
30 structure 720 and a laterally narrower center portion of the composite structure 720. In the embodiment illustrated in FIG. 7D the fibers 722 are arranged longitudinally along the composite structure 720. In other embodiments, at least some of the fibers 722 may be arranged laterally across the composite structure 720.

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Embodiments of the disclosure facilitate forming tooling for an EFAS system from composite materials. Forming the tooling from composite materials may facilitate tooling having higher density and/or higher graphitization. Forming the tooling from composite materials may also facilitate improved control over the electrical and thermal properties of the tooling, such as controlling where heat is generated and reducing the generation of heat in areas where heating is not needed.

Controlling the electrical and thermal properties of the tooling may reduce the power used by the EFAS system during an EFAS process. For example, controlling the electrical and thermal properties of the tooling may cause less heat to be generated in regions that are not adjacent to the part being formed. This may reduce the power used to generate the heat in the tooling. In other examples, concentrating the heat in a region proximate the part being formed may reduce an amount of time for the part to reach the desired elevated temperature, which may in turn reduce the processing time. Reducing the processing time may similarly reduce the power used to generate the heat for the EFAS processes.

Non-limiting example embodiments of the disclosure include:

Embodiment 1: A method of forming a composite, comprising: forming a composite preform through additive manufacturing; pyrolyzing the composite preform to form a porous composite preform; infiltrating the porous composite preform with a carbon precursor to form an infiltrated composite preform; carbonizing the infiltrated composite preform to form a carbonized composite preform; and exposing the carbonized composite preform to electric field assisted sintering (EFAS).

Embodiment 2: The method of embodiment 1, wherein infiltrating the porous composite preform with the carbon precursor comprises infiltrating the porous composite preform with mesophase pitch.

Embodiment 3: The method of embodiment 1 or embodiment 2, wherein infiltrating the porous composite preform with the carbon precursor comprises infiltrating the porous composite preform with at least one of a naphthalene-based mesophase pitch or an equivalent carbon heavy resin.

Embodiment 4: The method of any one or embodiments 1 through 3, wherein infiltrating the porous composite preform with the carbon precursor comprises infiltrating the porous composite preform with mesophase pitch and carbon black.

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Embodiment 5: The method of any one of embodiments 1 through 4, wherein exposing the carbonized composite preform to the EFAS comprises exposing the carbonized composite preform to heat and pressure to tailor a density of a carbon-carbon composite.

5 Embodiment 6: The method of any one of embodiments 1 through 5, wherein exposing the carbonized composite preform to the EFAS comprises graphitizing the carbonized carbon fiber preform.

 Embodiment 7: The method of any one of embodiments 1 through 6, wherein forming the composite preform through additive manufacturing comprises controlling an
10 orientation of individual fibers in the composite preform.

 Embodiment 8: The method of any one of embodiments 1 through 7, further comprising repairing the composite by exposing the composite to electric field assisted sintering (EFAS).

 Embodiment 9: A composite structure, comprising: carbon fibers embedded in a
15 graphitized matrix and comprising; a first region wherein the carbon fibers are oriented in a longitudinal orientation parallel to an axis of the composite structure; and a second region wherein the carbon fibers are oriented in a lateral orientation perpendicular to the axis of the composite structure.

 Embodiment 10: The composite structure of embodiment 9, wherein the composite
20 structure exhibits a density of from about 1.60 g/cm³ to about 2.0 g/cm³.

 Embodiment 11: The composite structure of embodiment 9 or embodiment 10, wherein the composite structure exhibits a diameter of from about 0.1 m to less than or equal to about 0.6 m.

 Embodiment 12: The composite structure of any one of embodiments 9 through 11,
25 wherein the composite structure is configured to be used as tooling in an electric field assisted sintering (EFAS) system.

 Embodiment 13: The composite structure of embodiment 12, wherein the second region comprises a high resistance region configured to generate heat when an electric current provided by the EFAS system passes through the composite structure.

30 Embodiment 14: The composite structure of embodiment 12 or embodiment 13, wherein the first region comprises a low resistance region configured to exhibit low power loss when an electric current provided by the EFAS system passes through the composite structure.

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Embodiment 15: The composite structure of any one of embodiments 9 through 14, wherein the first region is positioned at a different position along the axis of the composite structure from the second region.

Embodiment 16: The composite structure of any one of embodiments 9 through 15,
5 wherein the first region is positioned at a different distance radially from the axis of the composite structure from the second region.

Embodiment 17: A carbon-carbon composite, comprising: a continuous carbon fiber embedded in a graphitized matrix, the carbon-carbon composite exhibiting a density of from about 1.60 g/cm³ to about 2.0 g/cm³.

10 Embodiment 18: The carbon-carbon composite of embodiment 17, wherein the carbon-carbon composite exhibits a diameter of from about 0.1 m to less than or equal to about 0.6 m.

Embodiment 19: The carbon-carbon composite of embodiment 17 or
embodiment 18, wherein the carbon-carbon composite exhibits a degree of graphitization
15 in a range from about 50% to about 80%.

Embodiment 20: The carbon-carbon composite of any one of embodiments 17
through 19, further comprising: a first region wherein the continuous carbon fiber is
oriented in a longitudinal orientation parallel to an axis of the carbon-carbon composite;
and a second region wherein the continuous carbon fiber is oriented in a lateral orientation
20 perpendicular to the axis of the carbon-carbon composite.

Embodiment 21: The carbon-carbon composite of any one of embodiments 17
through 20, wherein the continuous carbon fiber is fused to adjacent carbon fibers forming
a bonded multidirectional network of reinforced fibers.

The embodiments of the disclosure described above and illustrated in the
25 accompanying drawing figures do not limit the scope of the invention, since these
embodiments are merely examples of embodiments of the invention, which is defined by
the appended claims and their legal equivalents. Any equivalent embodiments are intended
to be within the scope of this disclosure. Indeed, various modifications of the present
disclosure, in addition to those shown and described herein, such as alternative useful
30 combinations of the elements described, may become apparent to those skilled in the art
from the description. Such modifications and embodiments are also intended to fall within
the scope of the appended claims and their legal equivalents.

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CLAIMS

What is claimed is:

- 5 1. A method of forming a composite, comprising:
forming a composite preform through additive manufacturing;
pyrolyzing the composite preform to form a porous composite preform;
infiltrating the porous composite preform with a carbon precursor to form an infiltrated
composite preform;
10 carbonizing the infiltrated composite preform to form a carbonized composite preform; and
exposing the carbonized composite preform to electric field assisted sintering (EFAS).
2. The method of claim 1, wherein infiltrating the porous composite preform
with the carbon precursor comprises infiltrating the porous composite preform with
15 mesophase pitch.
3. The method of claim 1, wherein infiltrating the porous composite preform
with the carbon precursor comprises infiltrating the porous composite preform with at least
one of a naphthalene-based mesophase pitch or an equivalent carbon heavy resin.
20
4. The method of claim 1, wherein infiltrating the porous composite preform
with the carbon precursor comprises infiltrating the porous composite preform with
mesophase pitch and carbon black.
- 25 5. The method of claim 1, wherein exposing the carbonized composite preform
to the EFAS comprises exposing the carbonized composite preform to heat and pressure to
tailor a density of a carbon-carbon composite.
6. The method of claim 1, wherein exposing the carbonized composite preform
30 to the EFAS comprises graphitizing the carbonized carbon fiber preform.

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7. The method of any one of claims 1 through 6, wherein forming the composite preform through additive manufacturing comprises controlling an orientation of individual fibers in the composite preform.

5 8. The method of any one of claims 1 through 6, further comprising repairing the composite by exposing the composite to electric field assisted sintering (EFAS).

9. A composite structure, comprising:
carbon fibers embedded in a graphitized matrix and comprising;
10 a first region wherein the carbon fibers are oriented in a longitudinal orientation parallel to an axis of the composite structure; and
a second region wherein the carbon fibers are oriented in a lateral orientation perpendicular to the axis of the composite structure.

15 10. The composite structure of claim 9, wherein the composite structure exhibits a density of from about 1.60 g/cm³ to about 2.0 g/cm³.

11. The composite structure of claim 9, wherein the composite structure exhibits a diameter of from about 0.1 m to less than or equal to about 0.6 m.

20 12. The composite structure of any one of claims 9 through 11, wherein the composite structure is configured to be used as tooling in an electric field assisted sintering (EFAS) system.

25 13. The composite structure of claim 12, wherein the second region comprises a high resistance region configured to generate heat when an electric current provided by the EFAS system passes through the composite structure.

30 14. The composite structure of claim 12, wherein the first region comprises a low resistance region configured to exhibit low power loss when an electric current provided by the EFAS system passes through the composite structure.

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15. The composite structure of any one of claims 9 through 11, wherein the first region is positioned at a different position along the axis of the composite structure from the second region.

5 16. The composite structure of any one of claims 9 through 11, wherein the first region is positioned at a different distance radially from the axis of the composite structure from the second region.

17. A carbon-carbon composite, comprising:
10 a continuous carbon fiber embedded in a graphitized matrix, the carbon-carbon composite exhibiting a density of from about 1.60 g/cm³ to about 2.0 g/cm³.

18. The carbon-carbon composite of claim 17, wherein the carbon-carbon composite exhibits a diameter of from about 0.1 m to less than or equal to about 0.6 m.
15

19. The carbon-carbon composite of claim 17, wherein the carbon-carbon composite exhibits a degree of graphitization in a range from about 50% to about 80%.

20. The carbon-carbon composite of claim 17, further comprising:
20 a first region wherein the continuous carbon fiber is oriented in a longitudinal orientation parallel to an axis of the carbon-carbon composite; and
a second region wherein the continuous carbon fiber is oriented in a lateral orientation perpendicular to the axis of the carbon-carbon composite.

25 21. The carbon-carbon composite of any one of claims 17 through 20, wherein the continuous carbon fiber is fused to adjacent carbon fibers forming a bonded multidirectional network of reinforced fibers.

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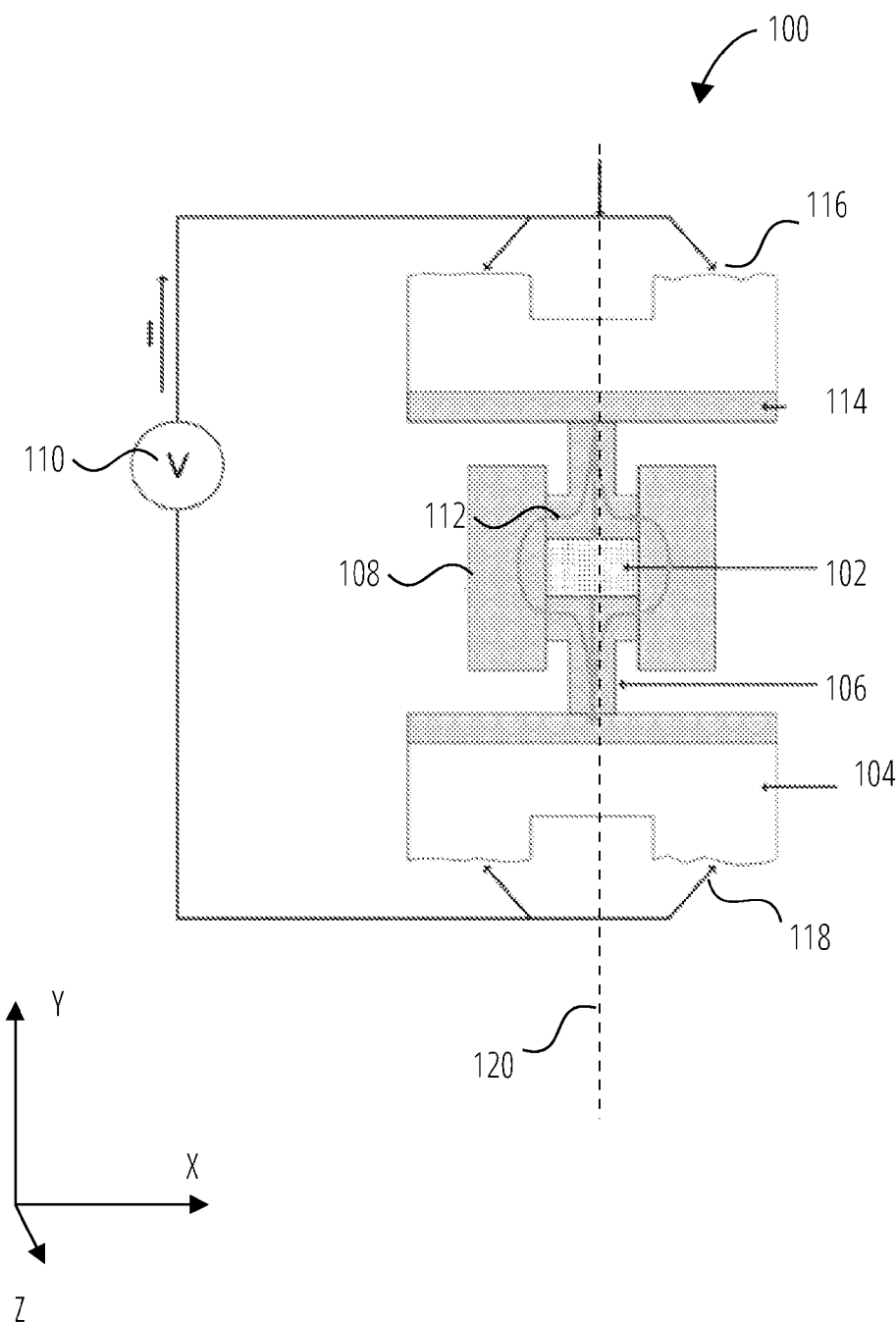


FIG. 1

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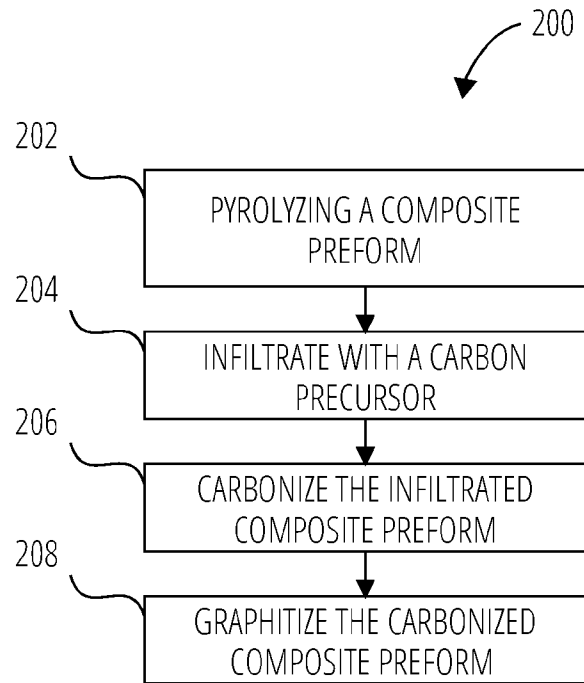
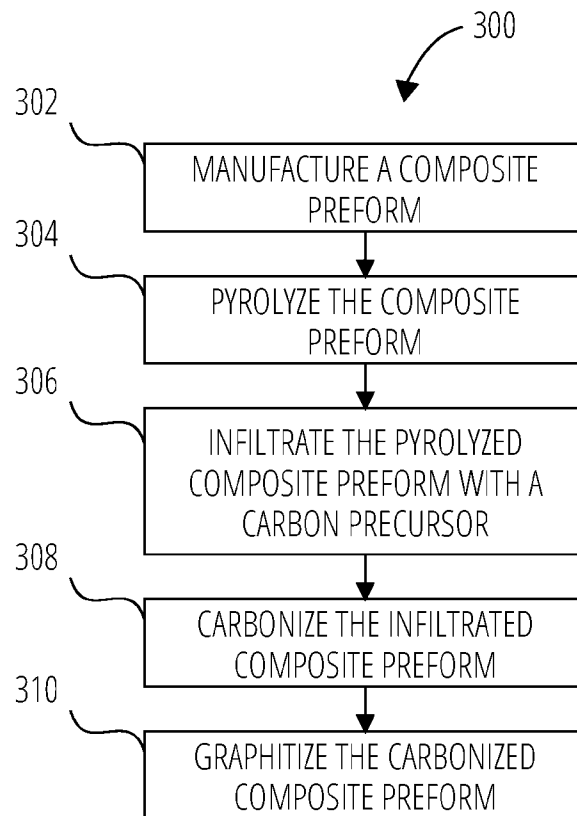


FIG. 2

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**FIG. 3**

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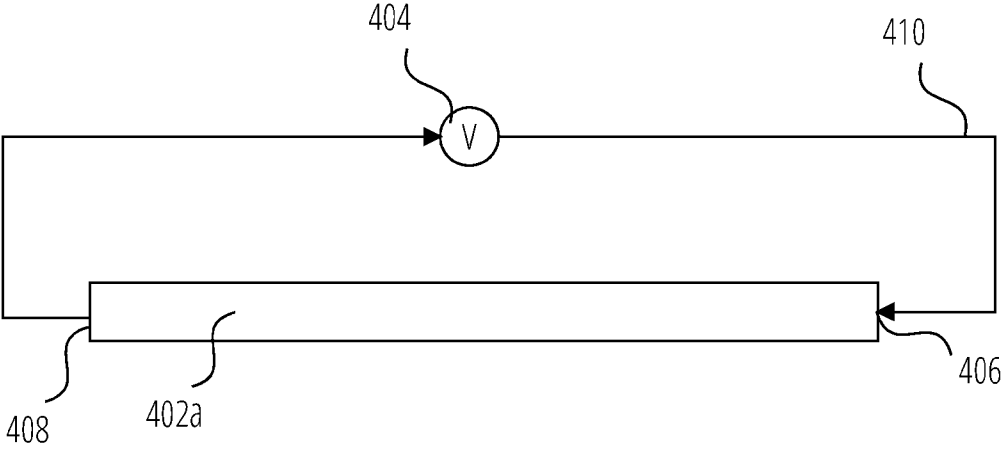


FIG. 4A

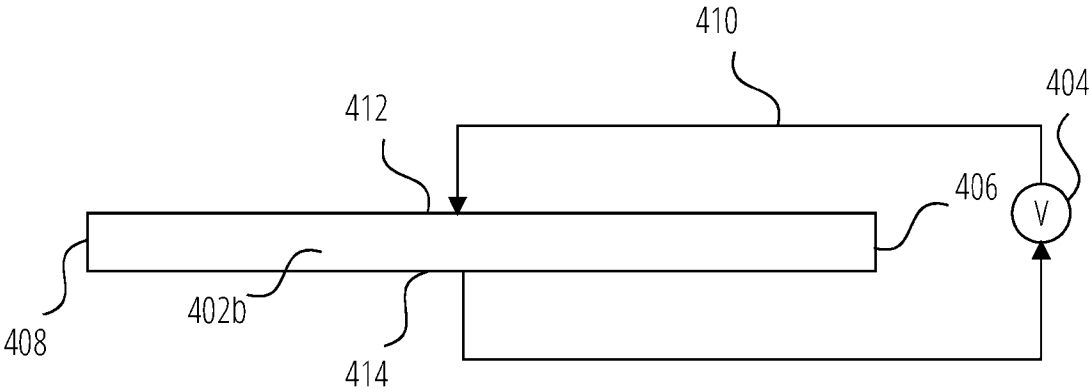


FIG. 4B

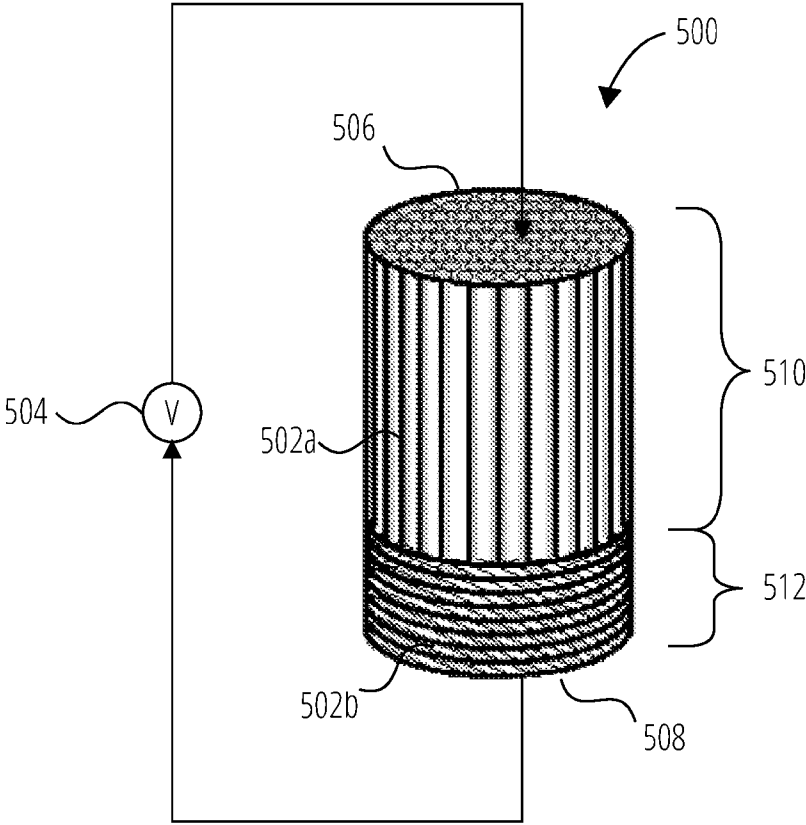


FIG. 5

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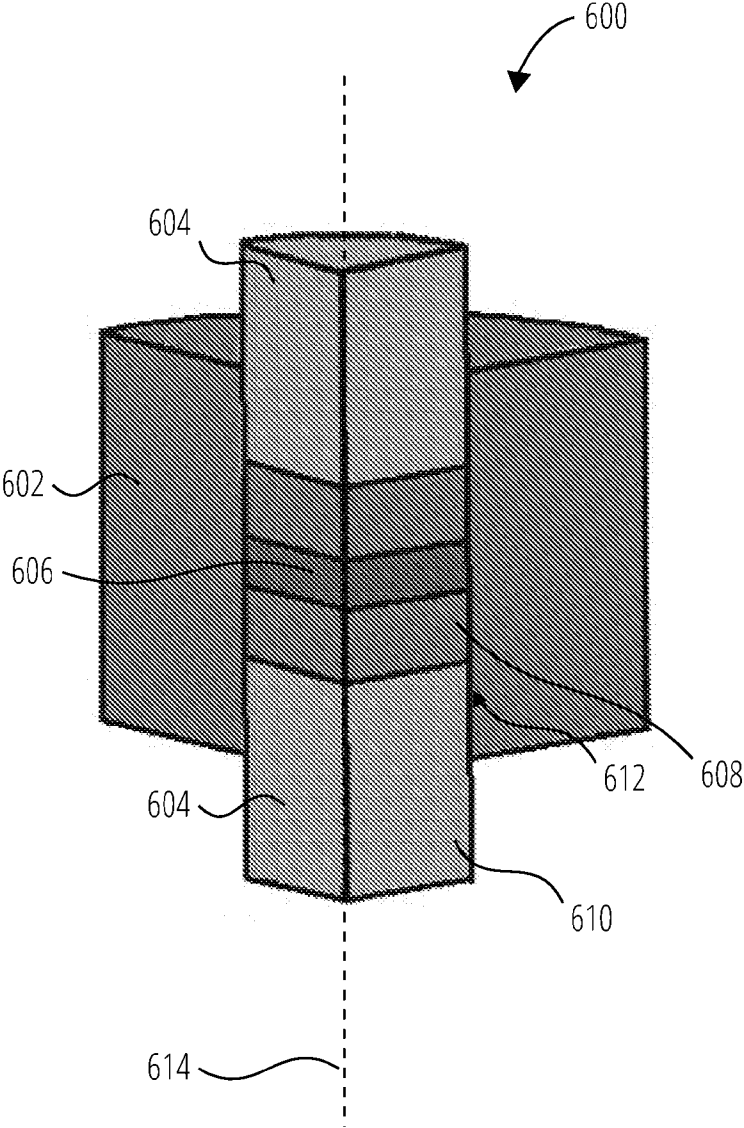


FIG. 6

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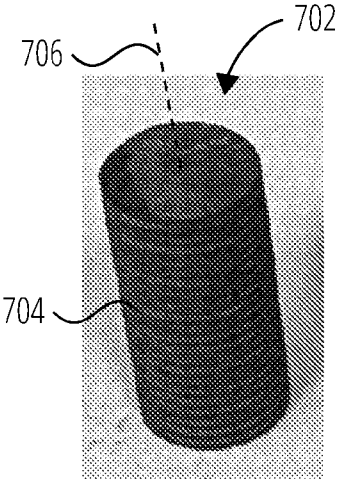


FIG. 7A

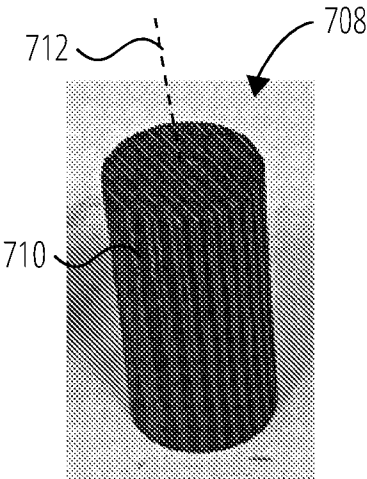


FIG. 7B

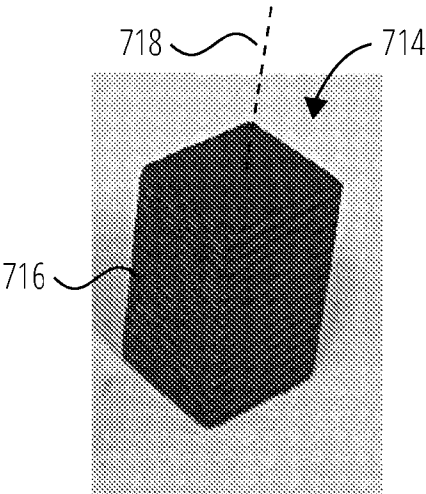


FIG. 7C

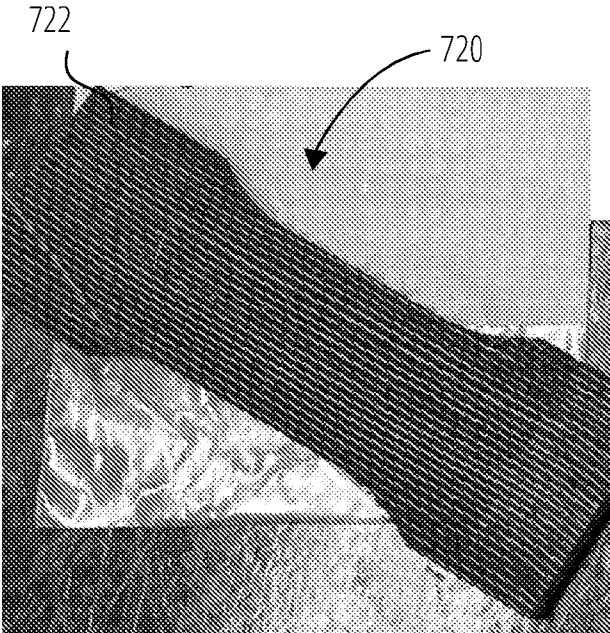


FIG. 7D