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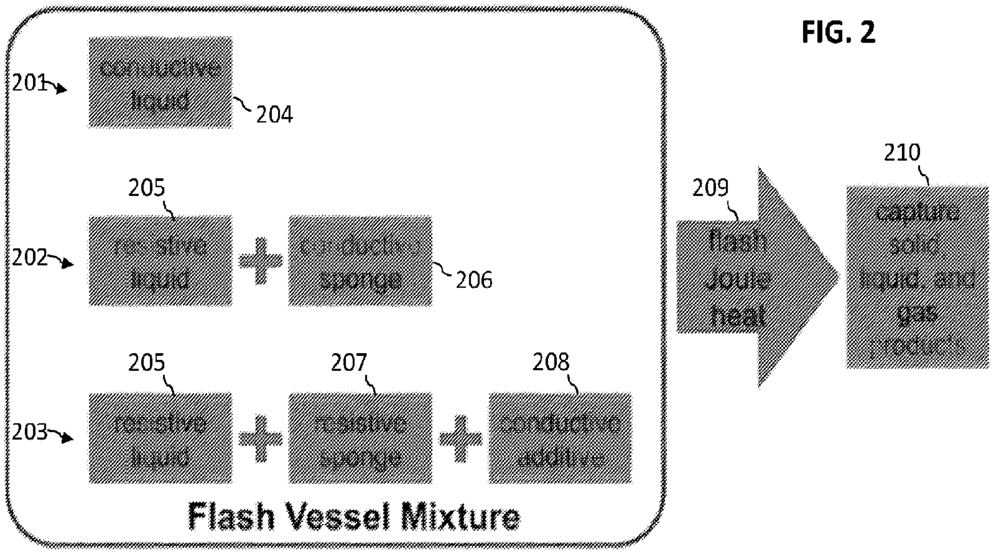

FIG. 2

(57) Abstract: Method and systems for flash Joule heating of liquids, particularly, methods and systems for flash Joule heating of liquids for gas capture, carbon/graphene templates (including customizable freestanding carbon/graphene templates), and carbon nanotubes.

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METHODS AND SYSTEMS OF FLASH JOULE HEATING OF LIQUIDS

CROSS-REFERENCE TO RELATED PATENT APPLICATIONS

[0001] The application claims priority to: (1) U.S. Patent Appl. Serial No. 63/595,646, filed November 2, 2023, entitled "Synthesis Of Hydrogen Gas From Waste Plastic Materials By Flash Joule Heating," and (2) U.S. Patent Appl. Serial No. 63/565,178, filed March 14, 2024, entitled "Methods And Systems Of Flash Joule Heating Of Liquids."

[0002] The methods and system of the present invention are also related to the PCT Patent Appl. Serial No. PCT/US24/48588, to James M. Tour, *et al.*, filed on September 26, 2024, entitled "Ultrafast Flash Joule Heating Synthesis Methods and Systems For Performing Same" ("*the Tour '588 PCT Application*").

[0003] The methods and system of the present invention are also related to the PCT Patent Appl. Serial No. PCT/US24/19867, to James M. Tour, *et al.*, filed on March 14, 2024, entitled "Synthesis Of Hydrogen Gas By Flash Joule Heating" ("*the Tour '867 PCT Application*").

[0004] The methods and systems of the present invention are also related to PCT Patent Appl. Serial Nos. PCT/US223/67000 to James M. Tour *et al.,* filed May 15, 2023, entitled "Flash Joule Heating For Production Of 1d Carbon And/Or Boron Nitride Nanomaterials" ("*the Tour '000 PCT Application*").

[0005] The methods and systems of the present invention are also related to PCT Patent Appl. Serial Nos. PCT/US21/52030, PCT/US21/52043, PCT/US21/52057, and PCT/US21/52070, to James M. Tour *et al.,* each entitled "Ultrafast Flash Joule Heating Synthesis Methods And Systems For Performing Same," each filed September 24, 2021 (collectively "*the Tour '070 PCT Applications*").

[0006] The methods and systems of the present invention are also related to U.S. Patent Appl. Serial No. 17/272,895, to James M. Tour, *et al.*, filed on March 2, 2021, entitled "Flash Joule Heating Synthesis Method And Compositions Thereof" ("*the Tour '895 Application*").

[0007] Each of these patent applications is commonly owned by the owner of the present invention and is incorporated herein in its entirety.

TECHNICAL FIELD

[0008] The present invention relates to methods and systems for flash Joule heating of liquids, particularly, methods and systems for flash Joule heating of liquids for gas capture, carbon/graphene templates (including customizable freestanding carbon/graphene templates, and carbon nanotubes.

GOVERNMENT INTEREST

[0009] This invention was made with government support under Grant No. FA9550-22-1-0526, awarded by the United States Air Force Office of Scientific Research, and Grant No. W912HZ-21-2-0050, awarded by the United States Army Corps of Engineers, Engineer Research and Development Center. The United States government has certain rights in the invention.

BACKGROUND

[0010] Flash Joule heating is a technique used to rapidly heat solid feedstocks through Joule heating, in which electric current flows through the target sample, in order to chemically convert the sample. Most commonly, this technique is used on amorphous carbon feedstocks to convert the carbon into graphene, as first published in 2020. [*Luong 2020*] This technique enables waste carbon sources, such as plastic waste, [*Wyss 2020*] to be upcycled to higher value graphene. The gases produced by this conversion process, such as alkane gases and hydrogen, can also be captured as products. [*Deng 2022*]. Flash Joule heating has also been used to synthesize doped graphene [*Chen 2022*] and carbon nanotubes [*Wyss 2023*] by mixing catalysts or doping agents into the reaction feedstock. However, flash Joule heating has previously been limited only to solid feedstocks.

SUMMARY OF INVENTION

[0011] The present invention relates to methods and systems for flash Joule heating of liquids, particularly, methods and systems for flash Joule heating of liquids for gas capture, carbon/graphene templates (including customizable freestanding carbon/graphene templates), and carbon nanotubes. The flash Joule heating of liquid provides an upcycling path to turn waste liquids into high-value graphene, carbon nanotubes, alkane gases, and hydrogen gas.

[0012] In general embodiments, the present invention is directed to a method that includes selecting a liquid feedstock. The method further includes performing a flash Joule heating process utilizing the liquid feedstock by applying a voltage or voltage pulse across the liquid feedstock. The feedstock is flash Joule heated.

[0013] Implementations of the invention can include one or more of the following features:

[0014] The liquid feedstock is flash Joule heated performing a high-temperature flash Joule heating process at a temperature of at least 2000 K.

[0015] The liquid feedstock is flash Joule heated at a temperature between 1000 and 1500 K to form a product comprising graphene.

[0016] The liquid feedstock is flash Joule heated at a temperature that is at most500 K to form a product comprising carbon nanotubes.

[0017] The flash Joule heating process can be performed by applying a voltage across the liquid feedstock.

[0018] The flash Joule heating process can be performed by applying a voltage pulse across the liquid feedstock.

[0019] The Joule heat process can heat the liquid feedstock and can convert the liquid feedstock to form one or more fluid or solid products.

[0020] The one or more fluid or solid products can include a fluid product that is a gas.

[0021] The one or more fluid or solid product can include a solid product selected from the group consisting of graphene, carbon/graphene templates, and nanotubes.

[0022] The liquid feedstock can be a liquid carbon source feedstock.

[0023] The method can further include upcycling the liquid carbon source into a product selected from the group consisting of doped and non-doped graphene and carbon nanotubes.

[0024] The method can further include making templates including carbon and/or graphene.

[0025] The template can be a customizable freestanding carbon and/or graphene template.

[0026] The step of making the template including carbon and/or graphene can include making a moldable carbon and graphene sponge for templated carbon nanotube growth and/or bottle-brush carbon nanotube growth.

[0027] The flash Joule heat process can heat the liquid feedstock and convert the liquid feedstock to form a gas product. The method can further include capturing the gas product.

[0028] The gas product can include a gas selected from the group consisting of carbon-based gases and dihydrogen (H_2).

[0029] The gas product can include a gas selected from the group consisting of carbon-based gases that are alkanes.

[0030] The gas product can be not inclusive of CO_2.

[0031] The flash Joule heating process can be performed in an inert atmosphere.

[0032] The flash Joule heating process can be performed in an argon or nitrogen atmosphere.

[0033] The performing of the flash Joule heating process utilizing the liquid feedstock can be performed with a mixture of the liquid feedstock and a catalyst and/or a doping agent.

[0034] The performing of the flash Joule heating process utilizing the liquid feedstock can be performed in combination with a catalyst including ferrocene.

[0035] The flash Joule heat process can heat the liquid feedstock and can convert the liquid feedstock to boron nitride nanotubes or transition metal dichalcogenide nanotubes.

[0036] The flash Joule heat process can heat the liquid feedstock and can convert the liquid feedstock to transition metal dichalcogenide nanotubes selected from the group consisting of MoS_2, $MoSe_2$, WS_2, WSe_2, TiO_2, TiS_2, $TiSe_2$, and combinations thereof on carbon nanotubes.

[0037] The liquid feedstock can include a liquid selected from the group consisting of oil, olive oil, paraffin oil, crude oil, motor oil, waste motor oil, waste oil, waste cooking oil, and combinations thereof.

[0038] The liquid feedstock can include a liquid selected from pitch, tar, wax, bitumen, crude oil, asphalt, asphaltene, oil sands, coal tar, biologically derived liquids, solids that have melted upon heating at a temperature below 1000 K, proteins, carbohydrates, and oils.

[0039] The liquid feedstock can be a viscous liquid.

[0040] The step of selecting a liquid feedstock can include selecting a solid material that melts into a liquid at a temperature below 1000 K.

[0041] The viscous liquid or solid material can be melted at a temperature below 1000 K.

[0042] The melting can be performed before the flash Joule heating process.

[0043] The melting can be performed utilizing a low-temperature flash Joule heating process at a temperature that is at most 1000 K.

[0044] The melted liquid can be melted into a sponge before the flash Joule heating.

[0045] The sponge can be selected from the group consisting of whole or chopped carbon fiber sponges and whole or chopped carbon nanotube sponges.

[0046] The liquid feedstock can be a conductive liquid.

[0047] The liquid feedstock can be mixed with a conductive additive.

[0048] The step of performing the flash Joule heating process utilizing the liquid feedstock can be performed without the liquid feedstock being sorbed on a sponge.

[0049] The method can further include sorbing the liquid feedstock on a sponge before the step of performing the flash Joule heating process. The step of performing the flash Joule heating process can include utilizing the liquid feedstock sorbed on the sponge.

[0050] The liquid feedstock can be a resistive liquid.

[0051] The sponge can be a conductive sponge.

[0052] The sponge can be a resistive sponge. A conductive additive can be added to the sponge and the liquid feedstock.

[0053] The sponge can be selected from the group consisting of carbon-based sponges, cellulose-based sponges, glass-based sponges, glass wool, ceramic-based sponges, and combinations thereof.

[0054] The method can produce a composite material.

[0055] In further general embodiments, the present invention is directed to a system that includes a liquid feedstock source and a reactor for performing flash Joule heating.

[0056] Implementations of the invention can include one or more of the following features:

[0057] The system can be operable to perform one or more of the above-described methods.

BRIEF DESCRIPTION OF THE DRAWINGS

[0058] **FIG. 1** shows a scheme of the synthesis enabled by flash Joule heating of liquids.

[0059] **FIG. 2** shows a scheme of the liquid flashing process. A general workflow for the synthesis process of flash Joule heating of liquids is shown.

[0060] **FIGS. 3A-3B** show two liquid flash vessels. **FIG. 3A** shows a conductive liquid (saltwater) inside a fused quartz flashing vessel. The flashing vessel is capped at either end with graphite electrodes. This sample can be directly flash Joule heated. **FIG. 3B** shows a nonconductive liquid (paraffin oil) adsorbed onto metallurgical coke in a quartz tube flashing vessel.

[0061] **FIGS. 4A-4B** show reaction yield of flash Joule heating crude oil reactions as well as the use of different sponges. The y-axis is mass. The input mass is shown as well as the theoretical and actual yields. The theoretical yield is the maximum possible solid mass recoverable, calculated from the mass of carbon in the input. The actual yield is the mass measured at reaction completion. The liquid products were converted to gases and solid graphene. This final solid, graphene product was weighed to determine the actual yield. **FIG. 4A** shows the reaction mass yield can exceed >90% when the liquids are trapped well during the reaction, using metallurgical coke as a sponge, allowing for more complete conversion. **FIG. 4B** shows the results of different sponge combinations, including plastic foam, packing peanut, denim, paper towel, and metallurgical coke, for which the liquid was allowed to escape, and so the theoretical yield was not considered.

[0062] **FIGS. 5A-5B** show solid-liquid mixtures for liquid flashing. **FIG. 5A** shows 75% paraffin oil / 25% activated carbon. **FIG. 5B** shows 75% paraffin oil / 25% carbon black.

[0063] **FIG. 6** shows flash Joule heating reaction of metallurgical coke and oil in which the liquid and gas products are allowed to escape igniting in air during the flash.

[0064] **FIG. 7A** shows gas chromatography-mass spectroscopy results of the flash Joule heating of different oils.

[0065] **FIG. 7B** shows gas chromatography-thermal conductivity measurement results of the flash Joule heating using olive oil.

[0066] **FIG. 8** shows an image of the product of flash Joule heating pitch in a chopped carbon fiber matrix.

[0067] **FIG. 9** shows a scheme of flash liquid growth of carbon nanotubes (CNTs).

[0068] **FIG. 10** shows a scheme of flash liquid growth of bottlebrush CNTs.

[0069] **FIG. 11** shows moldable graphene sponges without any liquid.

[0070] **FIG. 12** shows temperature-current profile of felt 'R' reaction shown in **FIG. 11**.

[0071] **FIGS. 13A-13B** show Raman spectroscopy analysis of carbon felt-derived flash graphene sponge, the same material though not the same sample shown in **FIG. 11**. **FIG. 13A** shows comparison of carbon felt sponge with carbon felt sponge-derived flash graphene. **FIG. 13B** shows turbostratic (TS) peaks displayed by carbon felt flash graphene.

[0072] **FIG. 14** shows Raman spectroscopy analysis of carbon nanotube radial breathing modes grown on a carbon felt sponge.

[0073] **FIG. 15** shows an SEM image of raw carbon felt sponge.

[0074] **FIGS. 16A-16D** show SEM images of carbon nanotubes grown on a carbon felt graphene sponge template. This template is the same as that in **FIG. 14.**

[0075] **FIGS. 17A-17B** show, respectively, distribution of CNT radii and average diameter, as measured by SEM across of a section of the CNT on carbon felt graphene template sample. This carbon felt graphene template is the same as that from **FIG. 14.**

[0076] **FIG. 18** shows a schematic of a flash Joule heating vessel connected to gas capture vessel.

[0077] **FIG. 19** shows a gas vessel used in flash Joule heating of liquid reactants.

[0078] **FIG. 20** shows GC-MS data of gases trapped from the flash Joule heating reactions of both olive oil and crude oil, trapped in a carbon felt sponge, with metcoke conductive additive.

DETAILED DESCRIPTION

[0079] The present invention relates to methods and systems for flash Joule heating of liquids, particularly, methods and systems for flash Joule heating of liquids for gas capture, carbon/graphene templates (including customizable freestanding carbon/graphene templates), and carbon nanotubes.

[0080] Among other things, embodiments of the methods and systems can provide for: flash Joule heating liquid feedstocks; upcycle liquid carbon sources into doped and non-doped graphene and carbon nanotubes; make moldable carbon and graphene sponges for templated carbon nanotube growth and bottle-brush carbon nanotube growth; and capture the gaseous products from these methods and systems. As shown the schematic in **FIG. 1**, the flash Joule heating of liquids is enabled, and the solid and gaseous products are recovered. When carbon-based liquids, such as oils, are flashed, then the products are flash graphene, carbon-based gases, such as alkanes, and hydrogen gas. Liquids can also be used as a heteroatom dopant and catalyst delivery agent, facilitating the growth of doped graphene and doped or undoped carbon nanotubes and bottle-brush carbon nanotubes. Carbon felt can also be used as a sponge to absorb the liquid prior to flashing, making a moldable graphene sponge as the product. Both the reacted graphene sponge and the unreacted carbon sponge can also be used for the templated growth of carbon nanotubes. As used herein the term “sponge” refers to any sorbent material that can sorb the liquid feedstock. Further, herein, the terms “adsorbed,” “absorbed,” and “sorbed” can be used interchangeably. As used herein, the term “template” refers to the solid platform or substrate onto which materials (such as nanotubes) grow, and “templated” refers to the state of having been grown on a template.

[0081] **FIG. 2** shows a scheme of the liquid flashing process. The process chosen (processes **201-203**) depends on the conductivity of the feedstock. Solid, liquid, and gaseous products can be captured (in **210**) after the reaction **21** is completed. Parameters/declarations for flash Joule heating of liquids with gas capture and customizable freestanding graphene templates include:

[0082] The flash Joule heating of liquid feedstocks is possible either when the liquid itself is sufficiently conductive or when a conductive additive (conductive liquid **204**) is present (process **201**).

[0083] A sponge **206** may be incorporated into which the liquid **205** can absorb or adsorb to facilitate flashing. The liquid can be a resistive liquid, may, but is not required to, also be a conductive additive (process **202**).

[0084] A conductive additive **209** may be incorporated into the flashing vessel to facilitate flashing. The conductive additive **209** may be but is not required to also be a sponge (and also the sponge can be a resistive sponge **207** or a conductive sponge) (process **203**).

[0085] The net conductivity of the sample, as determined by the mixture of any combination of the liquid, sponge, and conductive additive, must be within a window appropriate for flash Joule heating. This can generally between 0.5-50 ohms between the two flashing electrodes.

[0086] When carbonaceous feedstock is used in the methods and systems as the liquid, sponge, or conductive additive, then the carbonaceous feedstock can be converted to flash graphene typically when the reaction reaches ~2500 K. If the feedstock is heated only to 1500 K or less, then generally lower conversion occurs. Other carbon sources can include pitch, tar, wax, bitumen, crude oil, asphalt, asphaltene, biologically derived liquids, solids that melt upon heating, proteins, carbohydrates, oils, *etc.*

[0087] Common carbonaceous liquid feedstocks that can be utilized are new or waste oils, including, but not limited to, olive oil, paraffin oil, crude oil, motor oil, waste motor oil, waste oil, and waste cooking oil.

[0088] Waste products can be used as the liquid, sponge, and conductive additive in order to upcycle them to flash graphene or nanotubes or graphite.

[0089] Liquids can be used as solvents and delivery agents to assist in carrying other chemicals, without the need for a more comprehensive mixing step. This facilitates the growth of carbon nanotubes and the heteroatom doping of graphene and carbon nanotubes, whether or not performed on carbon or graphene templates.

[0090] If the objective of the flash is to convert liquid into graphene or nanotubes, then a tight seal is generally required. If the objective of the flash is merely to flash a sample in spite of the presence of liquid, then gaps can (and generally should) be used at the edges of the tube to allow liquid and gas products to more easily escape.

[0091] Carbon sponges used can be converted to graphene and still keep their form, allowing customizable, moldable graphene shapes to be synthesized. The sponges used can also or additionally be cellulose-based sponges, glass-based sponges, glass wool, ceramic-based sponges, and combinations thereof. For instance, when based on glass are used, the product can include silicon carbide. *See Tour '588 PCT Application.*

[0092] When a catalyst, such as ferrocene, is added to the mixture, then carbon nanotubes (CNTs) will grow. When this occurs in the presence of an ordered carbon sponge, such as carbon felt, then the carbon felt acts as a template onto which the carbon nanotubes grow to form a bottlebrush-type material. At temperatures above approximately >2500 K, then the carbon felt is converted to graphene. Below this, the carbon felt is not converted. Carbon fiber (continuous or chopped) can be used in a similar manner to grow CNTs. The carbon source for growth of carbon nanotubes on carbon fiber may be either a liquid or a solid. Adding a catalyst, such as ferrocene, ferric chloride, nickel chloride, or cobalt oxide or chloride, can facilitate the nanotube growth. Boron nitride nanotubes can also be grown on this carbon fiber. The interface of these 1D materials may be doped, modified, sized, or functionalized through flash Joule heating. Other options for catalysts include metal, metalloid, transition metal, organometallic, polyoxometalate, *etc*. The metal may be either a cation, or in a complex cation, or an anion, or complex anion or an oxide form. Carbon nanotubes may be formed out of any of the carbonaceous feedstocks, whether liquid, conductive additive, or sponge/fiber template.

[0093] The resulting CNTs on carbon felt can be recursively used to grow bottle-brush CNTs using this method, which are CNTs grown onto CNTs. A CNT mesh without the carbon felt can also be used to grow bottle-brush CNTs. Flashing in the presence of a dopant can also be used to grow doped bottle-brush carbon nanotubes. This technique can also be used to grow boron nitride nanotubes, or transition metal dichalcogenide nanotubes, such as MoS_2, $MoSe_2$, WS_2, WSe_2, TiO_2, TiS_2, $TiSe_2$, *etc*., on carbon nanotubes.

[0094] Any gases released from this process can be captured, including alkane and hydrogen gases. The gaseous products captured are a function of the flashing power and feedstock type used. When carbon feedstocks are flashed, then higher power will produce higher portions of hydrogen in the gas, while lower power will produce higher portions of long-chain alkanes in the gas.

[0095] If the reaction is performed under an inert atmosphere, such as argon, and the feedstock has no elemental oxygen present, then no CO_2 is produced.

[0096] The methods and systems of the present invention can be used and complementary to existing methods that use solid feedstock. For example, the methods and systems of the present invention can be complementary to the methods and systems for synthesizing graphene by flash Joule heating disclosed and taught in the *Tour '895 Application*. Such methods and systems have been demonstrated with an unmodulated DC pulse, and a pulse width modulated DC electrical pulse from a capacitor bank discharge, and these can also be performed with modulated or non-modulated pulsed or continuous AC and DC current sources. Liquid flashing and gas capture has been achieved at a variety of scales, from 50 mg to 1 g. Gas capture has been achieved from 50 mg to 400 mg.

[0097] The ability to flash waste liquids and convert them into high value graphene and gases provides and upcycling opportunity for liquid waste streams.

<u>Flash Joule Heating of Liquids</u>

[0098] The methods and systems of the present invention builds on flash Joule heating by using solid feedstock previously described in patents and literature (such as Applicant's *Tour '000 PCT Application, the Tour '070 PCT Applications*, and *the Tour '895 Application*) to act as a conductive additive and/or a sponge for the liquid. In an embodiment, a procedure for flash Joule heating of liquids is described below.

[0099] If the liquid is conductive, then the liquid does not require any conductive additive. If it is sufficiently viscous within the flashing vessel, then no sponge is required. This is illustrated in **FIG. 3A**. (In **FIG. 3A**, the flashing vessel is capped at either end with graphite electrodes. This sample can be directly flash Joule heated. This simply illustrates that a liquid can be used).

- If the liquid is low in viscosity, then the liquid must first be sorbed by a "sponge", which can either sorb the liquid into its interior or onto its surface. A kitchen sponge, textile, carbon felt, activated carbon, or other porous substance can be used to sorb the liquid inside. Metallurgical coke is used as an inexpensive feedstock to sorb the liquid onto its surface.
- If the liquid is non-conductive, then a conductive additive is required. This can be the same as the additive used for the sponge, such as in the case of metallurgical coke, activated carbon, and carbon felt. **FIG. 3B** illustrates a liquid mixture where metallurgical coke is being used as both a sponge and a conductive additive. (The metallurgical coke acts as both a conductive additive and as an adsorptive sponge.)

[0100] This mixture is placed in a flashing vessel and capped at either side with electrodes. The ideal resistance of the whole vessel from electrode to electrode is 0.5-50 Ω. If the objective is merely to flash the solid in spite of the liquid, then porous or fritted electrodes are used to allow the liquid to escape. If the liquid is the target feedstock, then tight electrodes are used as shown in **FIGS. 3A-3B**.

[0101] The flashing vessel is then flash Joule heated multiple times with increasing power and energy. For a ~250 mg batch size, with a 1:1 liquid-solid mass ratio, the vessel was heated using a 0.624 F capacitor-based flash Joule heater, discharged at total of 3 times at 60 V, then 80 V, then 100 V, using a pulse-width modulated discharge with the consecutive duty cycle sequence of 10% for 1 s, 20 % for 0.5 s, and 50 % for 5 s.

- The resultant products were graphene when the reaction temperature reached or exceeded ~2500 K. The reaction mass yield was >90 % when the tight graphite electrodes was used and the objective is to target the liquid for the reaction, as seen in **FIG. 4A**. (**FIG. 4A** shows the reaction mass yield can exceed >90% when the liquids are trapped well during the reaction, allowing for more complete conversion. The theoretical yield is calculated from the case in which all the carbon in the reactants is converted to graphene, and all other atoms escape as gaseous products. The actual yield is the mass of the output, solid products.) When the objective is merely to flash in spite of the liquid and thus release the liquid from the chamber during the reaction through porous or fritted electrodes, then excess liquid is expelled during the reaction, as seen in **FIG. 4B**. (**FIG. 4B** shows the results of different sponge combinations for which the liquid was allowed to escape, and so the theoretical yield was not considered. The oil mass can exceed the sponge mass by several times the mass of the sponge).

- The mass of the liquid feedstock can exceed that of the solid feedstock and still be appropriate for a flash liquid reaction. However, the reaction is more effective when the liquid feedstock is absorbed or adsorbed to the solid feedstock. **FIGS. 5A-5B** (**FIG. 5A** shows 75% paraffin oil / 25% activated carbon. **FIG. 5B** shows 75% paraffin oil / 25% carbon black. Despite being present in the same mass ratio with respect to the liquid, carbon black more effectively absorbs paraffin oil than activated carbon.) Additional low-voltage flashes may be needed for higher liquid concentration.
- The heat of the reaction may combust some of the liquid feedstock and gaseous products formed, as seen in **FIG. 6** (showing flash Joule heating reaction of metallurgical coke and oil in which the liquid and gas products are allowed to

escape igniting in air during the flash).

[0102] The reaction is permitted to cool, and the resulting products are collected.

[0103] As noted above, liquid feedstocks that can be utilized are new or waste oils, including, but not limited to, olive oil, paraffin oil, crude oil, motor oil, waste motor oil, waste oil, and waste cooking oil. **FIG. 7A** shows gas chromatography-mass spectroscopy results of the flash Joule heating of motor oil (plot **701**), olive oil (plot **702**), and crude oil (plot **703**). **FIG. 7B** shows gas chromatography-thermal conductivity measurement results of the flash Joule heating used olive oil. (This product was flash Joule heated using an inexpensive commercial arc welder). The results of **FIG. 7B** show that when 200 mg of olive oil was absorbed into carbon felt and flash Joule heated, 17 mL of H_2 gas was produced along with trace amounts of methane and carbon monoxide gas. This gas can be collected, such as described below.

[0104] As also noted above, other carbon sources can be utilized that include pitch, tar, wax, bitumen, crude oil, asphalt, asphaltene, biologically derived liquids, solids that melt upon heating, proteins, carbohydrates, oils, *etc*. For example, the liquid feedstock can be viscous liquid, such as pitch or tar, or a petroleum solid that turns to a liquid with minimal heating, such as gilsonite, pyrene, naphthalene or other aromatic carbon species, which can be generally melted below 200 °C.

[0105] The process can include performing a low-temperature melting step (*i.e.*, ~1000 K or below) to melt the solid carbon source into the sponge before undergoing flash Joule heating, and/or by utilizing flash Joule heating for this low-temperature melting step. In embodiments, the sponge may also be whole or chopped carbon fiber.

[0106] Moreover, the carbon liquid and sponge flash reaction product can be used to make composites. For example, 30-70 wt% pitch may be melted into a 30-70 wt% carbon fiber matrix and then flash Joule heated to make a composite with high mechanical strength. As discussed below, the composites can also include carbon nanotubes if ~1 wt% ferrocene or other catalyst is mixed-in prior to flash Joule heating.

__Synthesis of Templated Carbon/Graphene Sponges and CNTs__

[0107] The methods and systems of the present invention utilize the flash Joule heating of liquids method to grow carbon nanotubes on carbon templates using liquid as a catalyst delivery method. In an embodiment, a procedure for flash Joule heating of liquids is described below and illustrated in the schemes of **FIGS. 9-10**. **FIG. 9** shows a scheme of flash liquid growth of carbon nanotubes (CNTs) using oil **901**, carbon felt sponge **902**, and catalyst **903**. When the reaction is performed at a low temperature **906** (~1000 K), then CNTs but not graphene is formed (CNTs on carbon felt template **907**). When the reaction is performed at a higher temperature **904** (> 2000 K), then graphene is formed (CNTs on graphene felt template **905**), and the yield of CNTs decreases in proportion to the flashing temperature. **FIG. 10** shows a scheme of flash liquid growth of bottlebrush CNTs **1007** using oil **1001**, CNT sponge **1002**, and catalyst **1003**. A low temperature reaction **1006** is generally preferred (~1000 K) to reduce the destruction of CNTs during flashing. As used herein, "bottlebrush" means a base polymer, with strands of other polymers (in this case also CNTs) emanating from the base polymer.

[0108] A catalyst is dissolved or mixed into a carbon liquid. This has been demonstrated with ferrocene into olive oil and paraffin oil. Other catalysts are possible such as nickel, molybdenum, or cobalt based complexes, sats or oxides.

- A dopant can be used in place of a catalyst to produce doped graphene. It can also be used in addition to the catalyst to produce doped CNTs.

[0109] Analogous to the general liquid flashing method, the liquid is placed into a reaction vessel with a carbon conductive additive and a carbon sponge. This was demonstrated with carbon felt and metallurgical coke.

[0110] The flashing vessel is then flash Joule heated multiple times with increasing power and energy. For a ~250 mg batch size, with a 1:1 liquid-solid mass ratio, the vessel was heated using a 0.624 F capacitor-based flash Joule heater, discharged at total of 2 times at 80 V, and then 100 V, using a pulse-width modulated discharge with the consecutive duty cycle sequence of 10% for 1 s, 20 % for 0.5 s, and 50 % for 5 s.

- The resultant products were CNTs grown on a carbon-felt derived moldable graphene template (**FIGS. 11-12**) when the temperature exceeds 2000 K. (In **FIG. 11**, a carbon felt 'R' was placed into the flash Joule heating vessel before flashing. The 'R' was completely converted into flash graphene and maintained its predominant shape. This same felt can be used as a template for CNT growth or as a liquid sponge to create moldable products of these reactions. **FIG. 12** shows temperature-current profile of felt 'R' reaction shown in **FIG. 11**, with plots **1201-1202** in **FIG. 12** showing current and temperature, respectively. The outer surface of the reaction reached above 1750 °C using a pulse-width modulated DC flash discharge.) When the reaction is performed at ~1000 K, then the products are CNTs grown on a carbon-felt template.
- CNTs can be used in place of the carbon felt in order to grow bottlebrush CNTs (CNTs grown onto CNTs).
- A heteroatom dopant can be used in place of or in addition to the catalyst to make heteroatom doped graphene or heteroatom doped carbon nanotubes, respectively.
- A boron and nitrogen feedstock can be used to grow boron nitride on the CNTs or carbon/graphene felt.

[0111] The reaction is permitted to cool, and the resulting products are collected.

Characterization of Moldable Carbon/Graphene Sponges and Templated CNTs

[0112] To characterize moldable carbon/graphene sponges, Raman spectroscopy analysis was the primary tool used to evaluate the product. To characterize templated CNTs, scanning electron microscopy (SEM) and Raman spectroscopy analysis were utilized.

[0113] The characterization for the conversion of moldable carbon sponges into graphene sponges is identical to the characterization of flash graphene. **FIG. 13A** (showing comparison of carbon felt sponge (plot **1301**) with carbon felt sponge-derived flash graphene (plot **1302**)); **FIG. 13B** (showing turbostratic (TS) peaks displayed by carbon felt flash graphene (plot **1311**)). Raman spectroscopy analysis shows a significant enhancement of the 2D peak at ~2700 cm^{-1} and furthermore the presence of TS_1 and TS_2 peaks in the carbon felt flash graphene indicate both its conversion to flash graphene and the turbostratic nature of this flash graphene.

[0114] Carbon nanotubes exhibit radial breathing modes identifiable by Raman spectroscopy analysis which both indicate the presence of carbon nanotubes and correspond to specific carbon nanotube configurations, described by carbon nanotube indices. **FIG. 14** (with plots **1401-1404** for experiment, 279 cm^{-1} (10,1), 328 cm^{-2} (9,0), and cumulative fit peak $R^2 = 0.82$, respectively) Carbon nanotubes grown on carbon felt or on carbon felt flash graphene templates will have the simultaneous features of both under Raman spectroscopy analysis. **FIG. 14** (product was made from flash Joule heating the carbon sponge, which absorbed a mixture of olive oil and ferrocene; the positions of the two peaks in **FIG. 14** indicated the carbon nanotube indices of the nanotubes present).

[0115] Direct observation of the CNTs and their growth on the carbon felt template is best performed via SEM. The carbon felt, which serves as a template for the CNTs, as seen in **FIG. 11** is composed of threads ~20 μm in diameter (**FIG. 15**). Carbon nanotubes can thus be grown on this template in which they adhere to the threads of the carbon felt (**FIGS. 16A-16D**). The diameter of these carbon nanotubes varies between 20 and 150 nm (**FIGS. 17A-17B**).

<u>**Gas Capture from Flash Joule Heating of Carbon Liquids**</u>

[0116] In embodiments of the present invention, the gas capture procedure can be performed as an optional addition to any of the other liquid procedures described in this disclosure. When the liquid feedstock has both carbon and hydrogen chemically present, such as in most oils, then the gaseous products of flashing are hydrogen gas and carbonaceous gases, such as methane, ethane or butane. If the reaction is performed under an inert environment, such as under argon, then the reactive elements present will only be those from the flash Joule heating feedstock. This means that the CO_2 concentration present in the captured gases is limited by the amount of oxygen in the reactants. When no oxygen is present in the oil, then no CO_2 is produced. In an embodiment, a procedure for the gas capture from oils is described below.

[0117] Select an oil, sponge, and conductive additive feedstock as described above for liquid flash Joule heating.

[0118] Place the reactants in a flashing vessel where one end of the vessel is porous so as to allow gas to escape and be captured in a separate vessel, as shown in the scheme of **FIG. 18**.

[0119] Perform the flash Joule heating reaction on the sample.

- Perform a faster, more intense reaction to increase the ratio of hydrogen and short chain carbon gases among the products. Perform a slower, less intense reaction to increase the ratio of long-chain carbon gases among the products.
- A typical flash Joule heating reaction to produce higher yield of hydrogen will involve flashing 200 mg of solid "sponge" and 50 mg of oil using a 59.2 mF capacitor-based flash Joule heater, flashing three times at 100 V, 120 V, and 140 V.
- A typical flash Joule heating reaction to produce higher yield of heavy gases will involve 200 mg of solid "sponge" and 50 mg of oil using a 113 mF system flashing three times at 80 V, 100 V, and 120 V. Increasing capacitance of the system moderately increases the flash duration and flash energy. Increasing the voltage of the system slightly increases the flash duration and significantly

increases the flash energy.

[0120] Close the gas capture vessel with the pipe switch as soon as the reaction finishes. The gases will then be trapped in the gas capture vessel, and the other products, such as graphene, will remain in the initial flashing vessel. An image of the gas capture vessel used is shown in **FIG. 19**.

[0121] The gases trapped in the gas capture vessel are comprised of argon from the inert flashing atmosphere as well as the gases formed during the flash Joule heating process. The composition of this trapped gas was evaluated by an Agilent 8890 gas chromatography system with 5977 B MSD and G4407 TCD, the latter of which is used to measure the hydrogen gas concentration. A GC chromatograph of gases trapped from two liquid flashes is exhibited in **FIG. 20** (with plots **2001-2002** for olive oil and crude oil, respectively).

[0122] While embodiments of the invention have been shown and described, modifications thereof can be made by one skilled in the art without departing from the spirit and teachings of the invention. The embodiments described and the examples provided herein are exemplary only, and are not intended to be limiting. Many variations and modifications of the invention disclosed herein are possible and are within the scope of the invention. The scope of protection is not limited by the description set out above, but is only limited by the claims which follow, that scope including all equivalents of the subject matter of the claims.

[0123] The disclosures of all patents, patent applications, and publications cited herein are hereby incorporated herein by reference in their entirety, to the extent that they provide exemplary, procedural, or other details supplementary to those set forth herein.

[0124] Amounts and other numerical data may be presented herein in a range format. It is to be understood that such range format is used merely for convenience and brevity and should be interpreted flexibly to include not only the numerical values explicitly recited as the limits of the range, but also to include all the individual numerical values or sub-ranges encompassed within that range as if each numerical value and sub-range is explicitly recited. For example, a numerical range of approximately 1 to approximately 4.5 should be interpreted to include not only the explicitly recited limits of 1 to approximately 4.5, but also to include individual numerals such as 2, 3, 4, and sub-ranges such as 1 to 3, 2 to 4, etc. The same principle applies to ranges reciting only one numerical value, such as “less than approximately 4.5,” which should be interpreted to include all of the above-recited values and ranges. Further, such an interpretation should apply regardless of the breadth of the range or the characteristic being described.

[0125] Unless defined otherwise, all technical and scientific terms used herein have the same meaning as commonly understood to one of ordinary skill in the art to which the presently disclosed subject matter belongs. Although any methods, devices, and materials similar or equivalent to those described herein can be used in the practice or testing of the presently disclosed subject matter, representative methods, devices, and materials are now described.

[0126] Following long-standing patent law convention, the terms “a” and “an” mean “one or more” when used in this application, including the claims.

[0127] Unless otherwise indicated, all numbers expressing quantities of ingredients, reaction conditions, and so forth used in the specification and claims are to be understood as being modified in all instances by the term “about.” Accordingly, unless indicated to the contrary, the numerical parameters set forth in this specification and attached claims are approximations that can vary depending upon the desired properties sought to be obtained by the presently disclosed subject matter.

[0128] As used herein, the term “about” and “substantially” when referring to a value or to an amount of mass, weight, time, volume, concentration or percentage is meant to encompass variations of in some embodiments ±20%, in some embodiments ±10%, in some embodiments ±5%, in some embodiments ±1%, in some embodiments ±0.5%, and in some embodiments

±0.1% from the specified amount, as such variations are appropriate to perform the disclosed method.

[0129] As used herein, the term “substantially perpendicular” and “substantially parallel” is meant to encompass variations of in some embodiments within ±10° of the perpendicular and parallel directions, respectively, in some embodiments within ±5° of the perpendicular and parallel directions, respectively, in some embodiments within ±1° of the perpendicular and parallel directions, respectively, and in some embodiments within ±0.5° of the perpendicular and parallel directions, respectively.

[0130] As used herein, the term “and/or” when used in the context of a listing of entities, refers to the entities being present singly or in combination. Thus, for example, the phrase “A, B, C, and/or D” includes A, B, C, and D individually, but also includes any and all combinations and subcombinations of A, B, C, and D.

REFERENCES

[0131] Chen, W., *et al.*, “Heteroatom-Doped Flash Graphene,” *ACS Nano*, **2022**, *16(4)*, 6646-6656 (“*Chen 2022*”).

[0132] Deng, B., *et al.*, “Phase controlled synthesis of transition metal carbide nanocrystals by ultrafast flash Joule heating,” *Nat. Commun.*, **2022**, *13(1)*, 262 (“*Deng 2022*”).

[0133] Luong, D. X., *et al.*, “Gram-scale bottom-up flash graphene synthesis,” *Nature*, **2020**, *577*, 647–651 (“*Luong 2020*”).

[0134] Wyss, K. M, *et al.*, “Upcycling Of Waste Products Into Hybrid Carbon Nanomaterials,” *Advanced Materials.*, **2023**, *35(16)*, 2209621 (“*Wyss 2023*”).

[0135] Wyss, K. M., *et al.*, “Converting Plastic Waste Pyrolysis Ash into Flash Graphene,” *Carbon*, **2021**, *174*, 430-438 (“*Wyss I 2021*”).

WHAT IS CLAIMED IS:

1. A method comprising:

(a) selecting a liquid feedstock; and

(b) performing a flash Joule heating process utilizing the liquid feedstock by applying a voltage or voltage pulse across the liquid feedstock, wherein the liquid feedstock is flash Joule heated.

2. The method of Claim 1, wherein the liquid feedstock is flash Joule heated performing a high-temperature flash Joule process at a temperature that is at least 2000 K.

3. The method of Claim 1, wherein the liquid feedstock is flash Joule heated at a temperature that is between 1000 K and 1500 K to form a product comprising graphene.

4. The method of Claim 1, wherein the liquid feedstock is flash Joule heated at a temperature that is at most 500 K to form a product comprising carbon nanotubes.

5. The method of Claim 1, wherein the Joule heat process heats the liquid feedstock and converts the liquid feedstock to form one or more fluid or solid products.

6. The method of Claim 5, wherein the one or more fluid or solid product comprises a solid product selected from the group consisting of graphene, carbon/graphene templates, and nanotubes.

7. The method of Claim 1, wherein the liquid feedstock is a liquid carbon source feedstock.

8. The method of Claim 7, wherein the method further comprises upcycling the liquid carbon source into a product selected from the group consisting of doped and non-doped graphene and carbon nanotubes.

9. The method of Claim 1, wherein the method further comprises making templates comprising carbon and/or graphene.

10. The method of Claim 1, wherein

(a) the flash Joule heat process heats the liquid feedstock and convert the liquid feedstock to form a gas product; and

(b) the method further comprises capturing the gas product.

11. The method of Claim 10, wherein the gas product comprises a gas selected from the group consisting of carbon-based gases and dihydrogen (H_2).

12. The method of Claim 1, wherein the flash Joule heating process is performed in an inert atmosphere.

13. The method of Claim 1, wherein the performing of the flash Joule heating process utilizing the liquid feedstock is performed with a mixture of the liquid feedstock and a catalyst and/or a doping agent.

14. The method of Claim 1, wherein the flash Joule heat process heats the liquid feedstock and converts the liquid feedstock to boron nitride nanotubes or transition metal dichalcogenide

nanotubes.

15. The method of Claim 1, wherein the liquid feedstock comprises a liquid selected from the group consisting of oil, olive oil, paraffin oil, crude oil, motor oil, waste motor oil, waste oil, waste cooking oil, and combinations thereof.

16. The method of Claim 1, wherein the liquid feedstock comprises a liquid selected from pitch, tar, wax, bitumen, crude oil, asphalt, asphaltene, oil sands, coal tar, biologically derived liquids, solids that have melted upon heating at a temperature below 1000 K, proteins, carbohydrates, and oils.

17. The method of Claim 1, wherein the liquid feedstock is a viscous liquid.

18. The method of Claim 1, wherein the step of selecting a liquid feedstock comprises selecting a solid material that melts into a liquid at a temperature below 1000 K.

19. The method of any of Claims 17-18, wherein the viscous liquid or solid material is melted at a temperature below 1000 K.

20. The method of Claim 19, wherein the melting is performed before the flash Joule heating process.

21. The method of Claim 19, where the melting is performed utilizing a low-temperature flash Joule heating process at a temperature that is at most 1000 K.

22. The method of any of Claims 19-21, wherein the melted liquid is melted into a sponge before the flash Joule heating.

23. The method of Claim 22, wherein the sponge is a selected from the group consisting of whole or chopped carbon fiber sponges and whole or chopped carbon nanotube sponges.

24. The method of any of Claims 1-23, wherein the liquid feedstock is a conductive liquid.

25. The method of any of Claims 1-23, wherein the liquid feedstock is mixed with a conductive additive.

26. The method of any of Claims 24-25, wherein the step of performing the flash Joule heating process utilizing the liquid feedstock is performed without the liquid feedstock being sorbed on a sponge.

27. The method of any of Claims 1-26, wherein

(a) the method further comprises sorbing the liquid feedstock on a sponge before the step of performing the flash Joule heating process; and

(b) the step of performing the flash Joule heating process comprises utilizing the liquid feedstock sorbed on the sponge.

28. The method of Claim 27, wherein the liquid feedstock is a resistive liquid.

29. The method of any of Claims 27-28, wherein the sponge is a conductive sponge.

30. The method any of Claim 27-28, wherein

(a) the sponge is a resistive sponge; and

(b) a conductive additive is added to the sponge and the liquid feedstock.

31. The method of Claim 27, wherein the sponge is selected from the group consisting of carbon-based sponges, cellulose-based sponges, glass-based sponges, glass wool, ceramic-based sponges, and combinations thereof.

32. The method of any of Claims 27-31, wherein the method produces a composite material.

33. A system that comprises a liquid feedstock source and a reactor for performing flash Joule heating.

34. The system of Claim 33, wherein where the system is operable for performing one or more of the methods of Claims 1-32.

WHAT IS CLAIMED IS:

1. A method comprising:

(a) selecting a liquid feedstock, wherein the liquid feedstock is a liquid carbon, boron, nitrogen, and/or silicon source feedstock that is in liquid form; and

(b) performing a flash Joule heating process utilizing the liquid feedstock by applying a voltage or voltage pulse across the liquid feedstock, wherein the liquid feedstock is flash Joule heated to convert the liquid carbon, boron, nitrogen, and/or silicon source feedstock to a product comprising 1-dimensional materials.

2. The method of Claim 1, wherein the liquid feedstock is flash Joule heated performing a high-temperature flash Joule process at a temperature that is at least 2000 K to form the product comprising the 1-dimensional materials.

3. The method of Claim 1, wherein the liquid feedstock is flash Joule heated at a temperature that is between 1000 K and 2500 K to form the product comprising the 1-dimensional materials.

4. The method of Claim 1, wherein the liquid feedstock is flash Joule heated at a temperature that is at most 2500 K to form the product comprising the 1-dimensional materials, wherein the 1-dimensational materials are carbon nanotubes.

5. The method of Claim 1, wherein the flash Joule heat process converts the liquid carbon, boron, nitrogen, and/or silicon source feedstock to form one or more fluid or solid 1-dimensional materials.

6. The method of Claim 5, wherein the one or more fluid or solid 1-dimensional materials comprises nanotubes.

7. The method of Claim 1, wherein the 1-dimensional materials are doped 1-dimensional materials.

8. The method of Claim 1, wherein the 1-dimensaton materials are non-doped 1-dimensional materials.

9. The method of Claim 1, wherein the method further comprising using templates silicon, nitrogen, carbon, and/or boron, or compounds thereof, as sponges to absorb the liquid carbon source feedstock.

10. The method of Claim 1, wherein

(a) the flash Joule heat process heats the liquid feedstock and convert the liquid feedstock to the product in form of a gas product; and

(b) the method further comprises capturing the gas product.

11. The method of Claim 10, wherein the gas product comprises a gas selected from the group consisting of carbon-based gases and dihydrogen (H_2).

12. The method of Claim 1, wherein the flash Joule heating process is performed in an inert atmosphere.

13. The method of Claim 1, wherein the performing of the flash Joule heating process

utilizing the liquid feedstock is performed with a mixture of the liquid feedstock and a catalyst and/or a doping agent.

14. The method of Claim 1, wherein the 1-dimensational materials comprise boron nitride nanotubes, silicon carbide nanowhiskers, and/or transition metal dichalcogenide nanotubes.

15. The method of Claim 1, wherein the liquid carbon source feedstock comprises a liquid selected from the group consisting of oil, olive oil, paraffin oil, crude oil, motor oil, waste motor oil, waste oil, waste cooking oil, and combinations thereof.

16. The method of Claim 1, wherein the liquid feedstock comprises a liquid selected from pitch, tar, wax, bitumen, crude oil, asphalt, asphaltene, oil sands, coal tar, biologically derived liquids, solids that have melted upon heating at a temperature below 1000 K, proteins, carbohydrates, and oils.

17. The method of Claim 1, wherein the liquid carbon, boron, nitrogen, and/or silicon source feedstock is a viscous liquid.

18. The method of Claim 1, wherein the step of selecting a liquid feedstock comprises selecting a solid material that is melted into the liquid form at a temperature below 1000 K.

19. The method of Claim 17, wherein the viscous liquid is melted at a temperature below 1000 K.

20. The method of any of Claims 18-19, wherein the melting is performed before the flash Joule heating process.

21. The method of any of Claims 18-19, where the melting is performed utilizing a low-temperature flash Joule heating process at a temperature that is at most 1000 K.

22. The method of any of Claims 18-21, wherein the melted liquid is melted into a sponge before the flash Joule heating.

23. The method of Claim 22, wherein the sponge is a selected from the group consisting of whole or chopped carbon fiber sponges and whole or chopped carbon nanotube sponges.

24. The method of any of Claims 1-23, wherein the liquid feedstock is an electrically conductive liquid.

25. The method of any of Claims 1-23, wherein the liquid feedstock is mixed with an electrically conductive additive.

26. The method of any of Claims 24-25, wherein the step of performing the flash Joule heating process utilizing the liquid feedstock is performed without the liquid feedstock being sorbed on a sponge.

27. The method of any of Claims 1-26, wherein

(a) the method further comprises sorbing the liquid feedstock on a sponge before the step of performing the flash Joule heating process; and

(b) the step of performing the flash Joule heating process comprises utilizing the liquid feedstock sorbed on the sponge.

28. The method of Claim 27, wherein the liquid feedstock is an electrically resistive liquid.

29. The method of any of Claims 27-28, wherein the sponge is an electrically conductive sponge.

30. The method any of Claim 27-28, wherein

(a) the sponge is an electrically resistive sponge; and

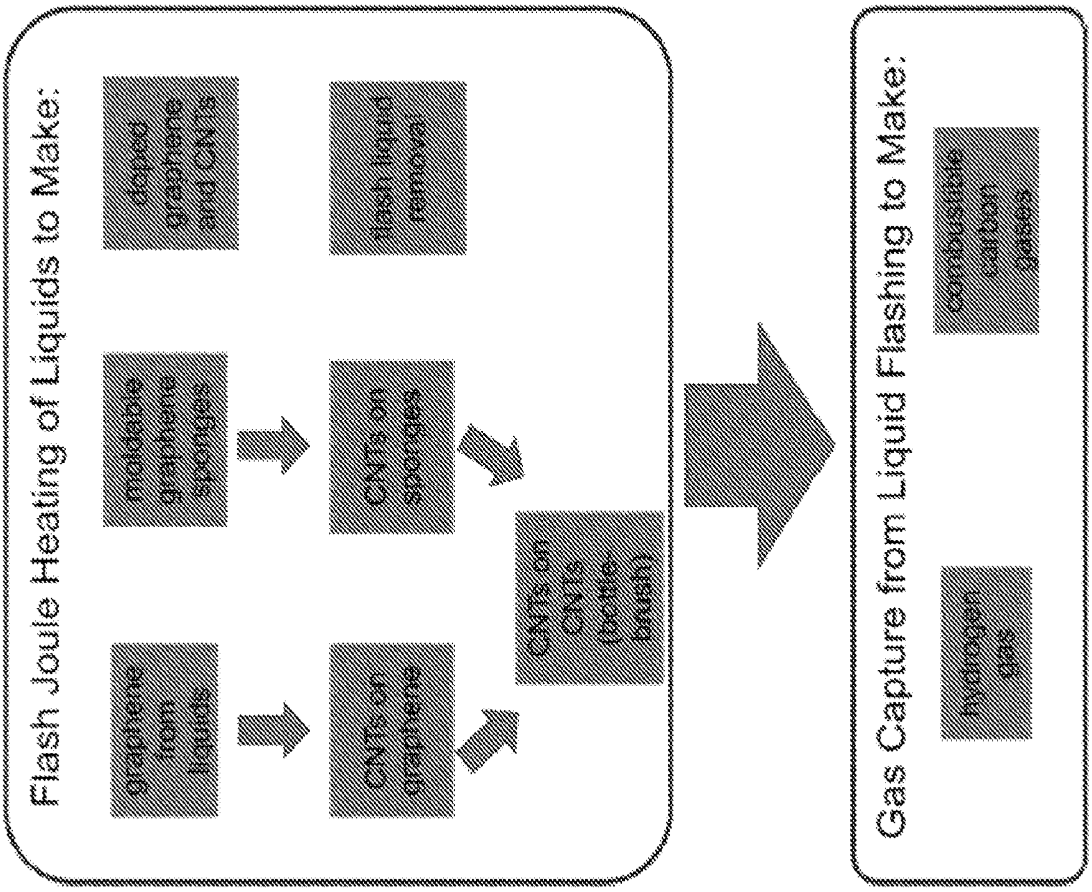
(b) an electrically conductive additive is added to the sponge and the liquid feedstock.

31. The method of Claim 27, wherein the sponge is selected from the group consisting of carbon-based sponges, cellulose-based sponges, glass-based sponges, glass wool, ceramic-based sponges, and combinations thereof.

32. The method of any of Claims 27-31, wherein the method produces a composite material which composite material consists of the carbon or carbide template material used as an electronically conductive additive, in which the liquid carbon-, boron-, nitrogen- and/or silicon-containing precursor is absorbed, bonded to the products generated from the liquid carbon-, boron-, nitrogen- and/or silicon-containing precursor during the flash Joule heating process.

33. A system that comprises a liquid feedstock source and a reactor for performing a flash Joule heating process operable for performing one or more of the methods of Claims 1-32.

34. (Cancelled).



Flash Joule Heating of Liquids to Make:
graphene from liquids
moldable graphene sponges
doped graphene and CNTs
CNTs on graphene
CNTs on sponges
flash liquid removal
CNTs on CNTs (bottle brush)
Gas Capture from Liquid Flashing to Make:
hydrogen gas
combustible carbon gases

FIG. 1

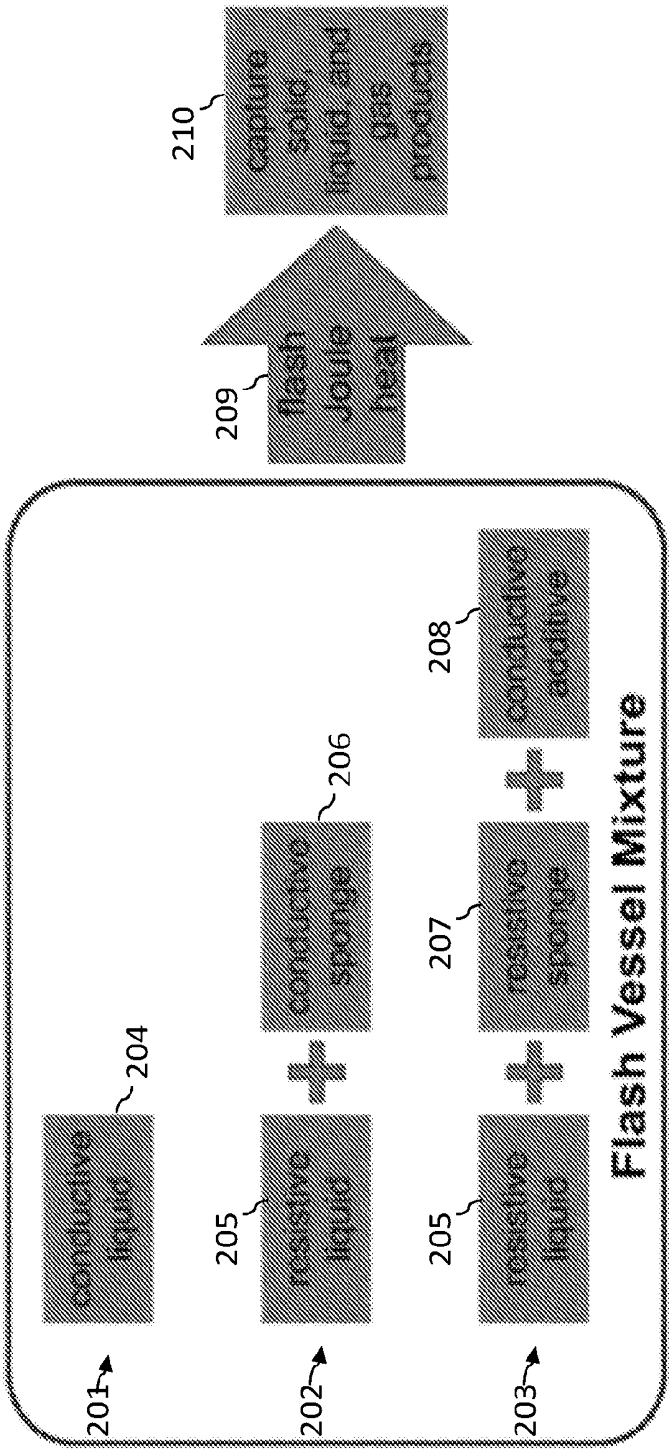

capture solid, liquid, and gas products
210
flash Joule heat
209
Flash Vessel Mixture
conductive liquid
204
201
resistive liquid
205
conductive sponge
206
202
resistive liquid
205
resistive sponge
207
conductive additive
208
203

FIG. 2

FIG. 3A

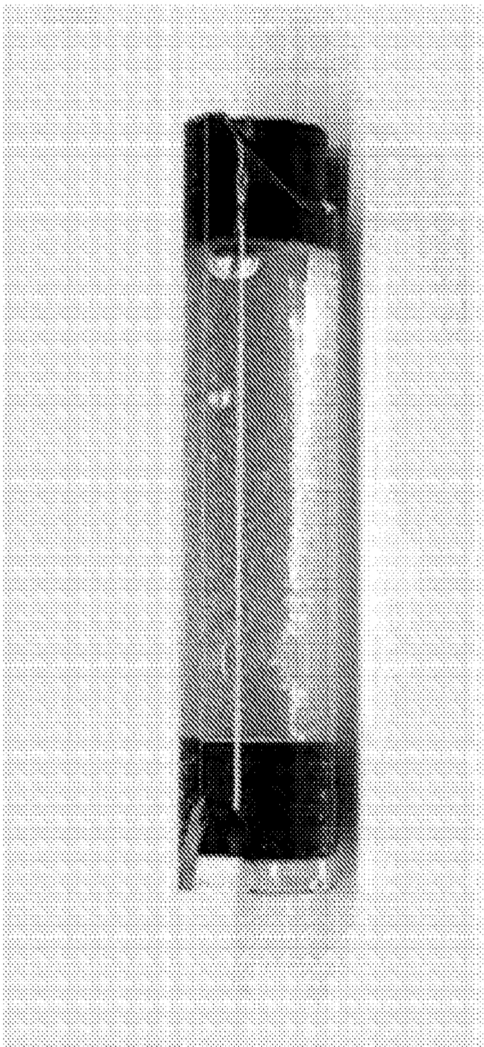
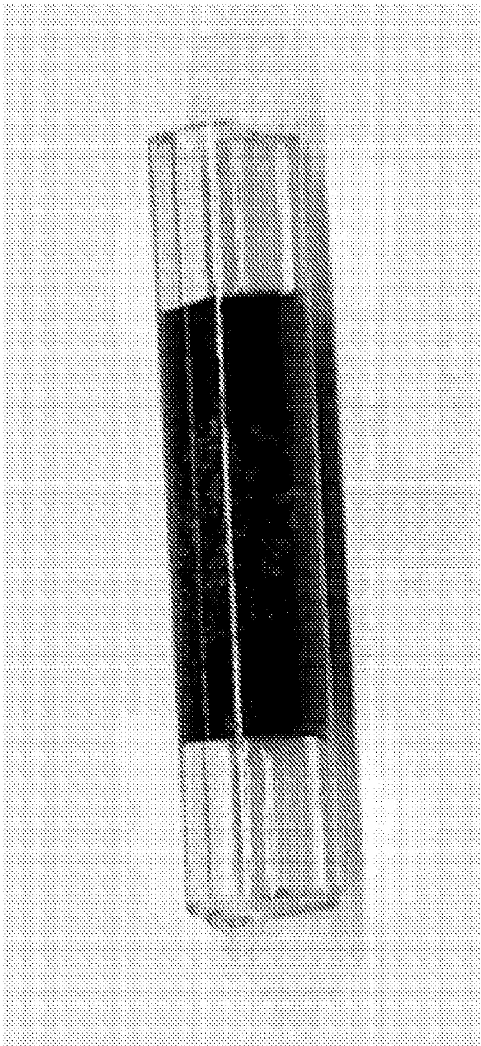


FIG. 3B



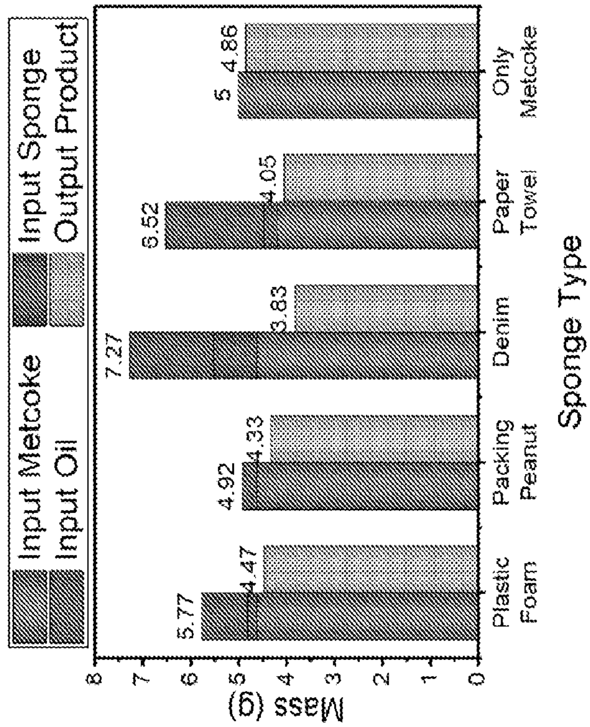

Input Metcoke
Input Oil
Input Sponge
Output Product
Mass (g)
8
7
6
5
4
3
2
1
0
5.77
4.47
4.92
4.33
7.27
3.83
6.52
4.05
5
4.86
Plastic Foam
Packing Peanut
Denim
Paper Towel
Only Metcoke
Sponge Type

FIG. 4B

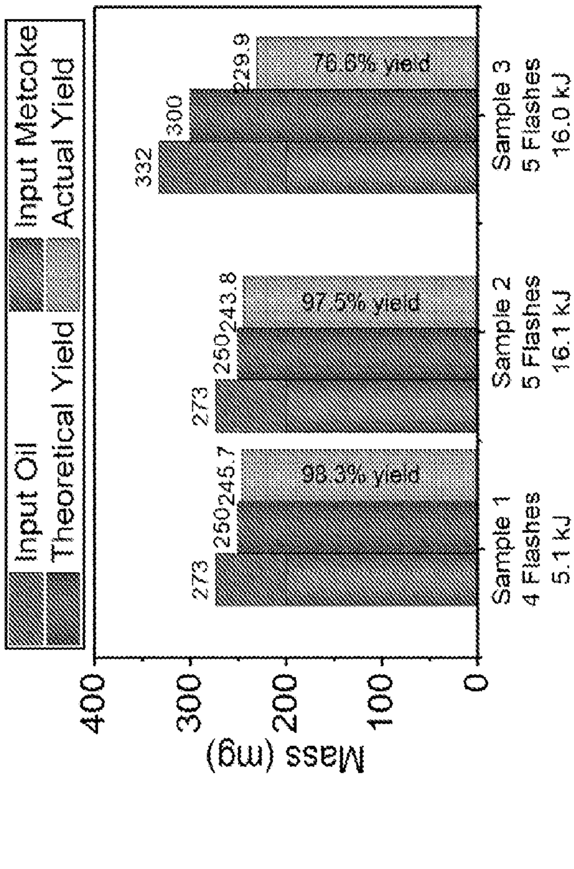

Input Oil
Theoretical Yield
Input Metcoke
Actual Yield
Mass (mg)
400
300
200
100
0
273
250
245.7
98.3% yield
273
250
243.8
97.5% yield
332
300
229.9
76.6% yield
Sample 1
4 Flashes
5.1 kJ
Sample 2
5 Flashes
16.1 kJ
Sample 3
5 Flashes
16.0 kJ

FIG. 4A

75 % Paraffin Oil
25 % Activated Carbon

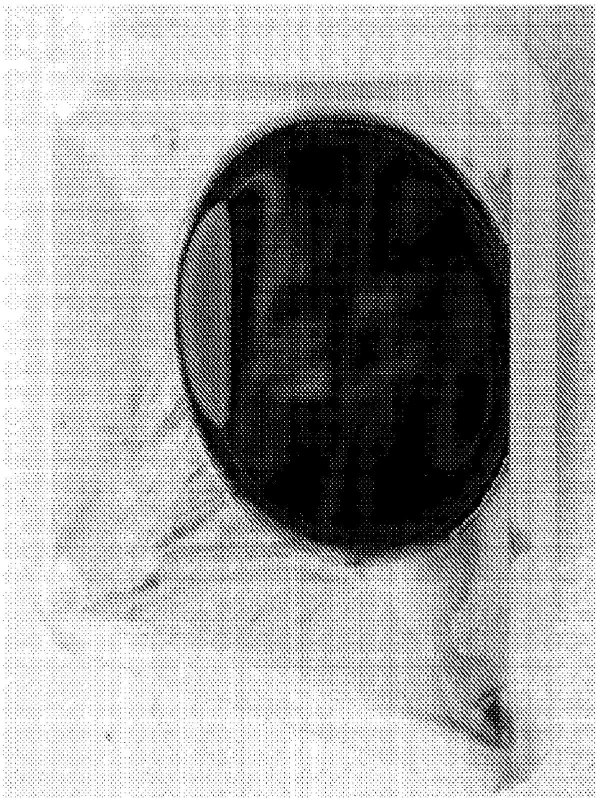


FIG. 5A

75 % Paraffin Oil
25 % Carbon Black

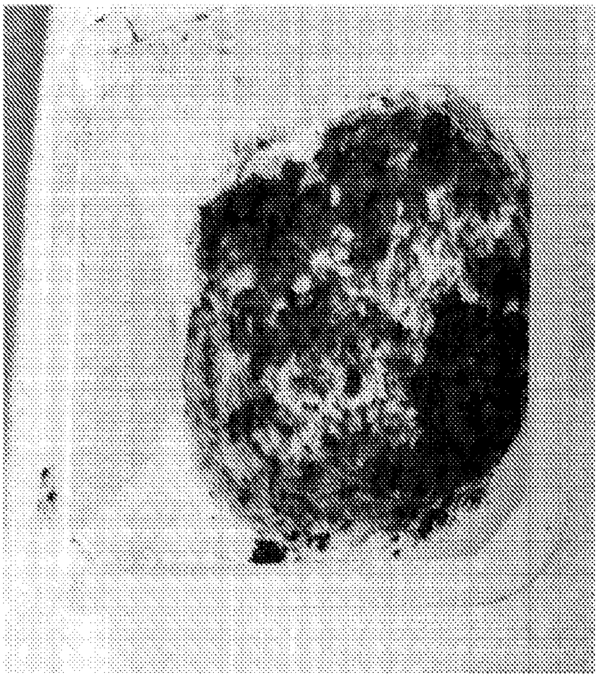


FIG. 5B

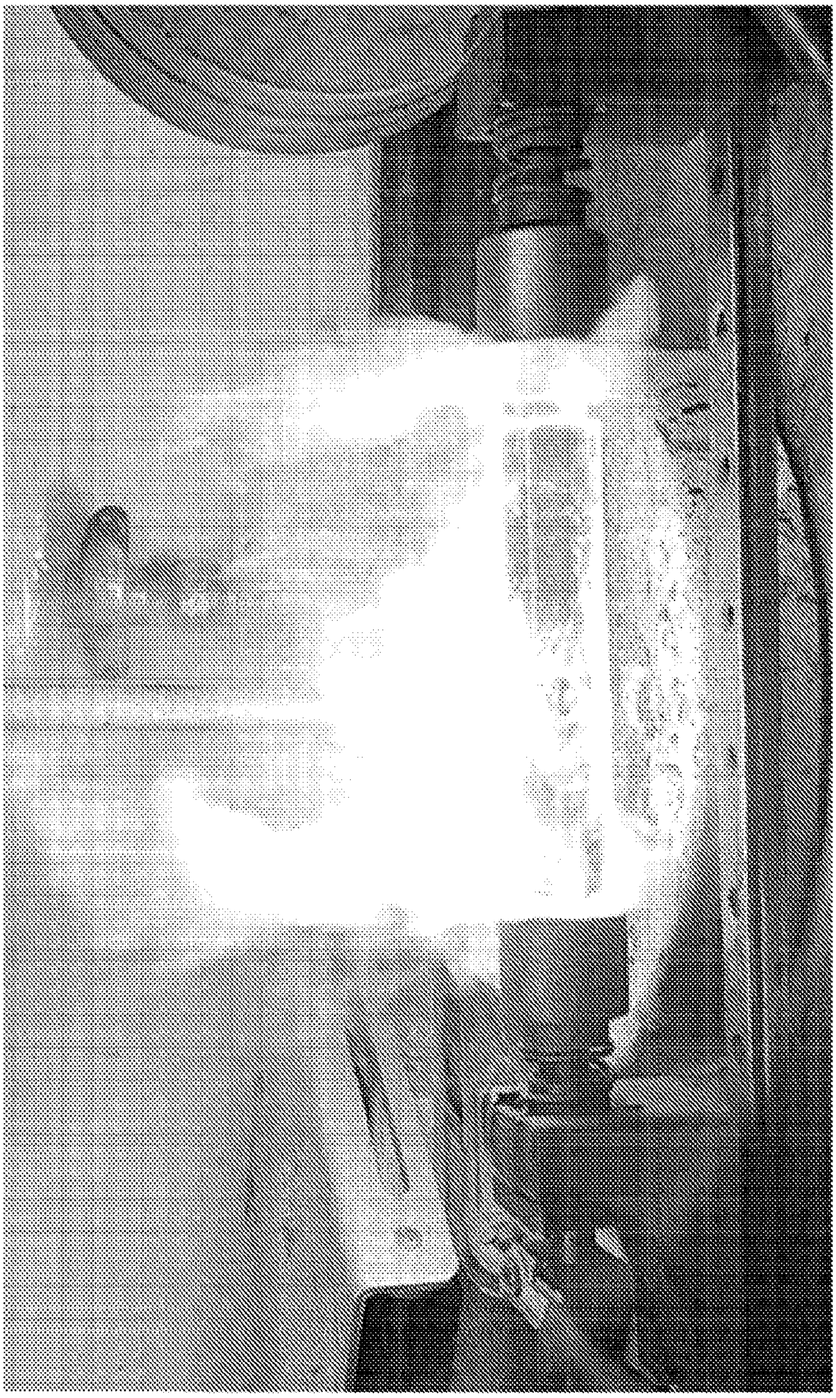


FIG. 6

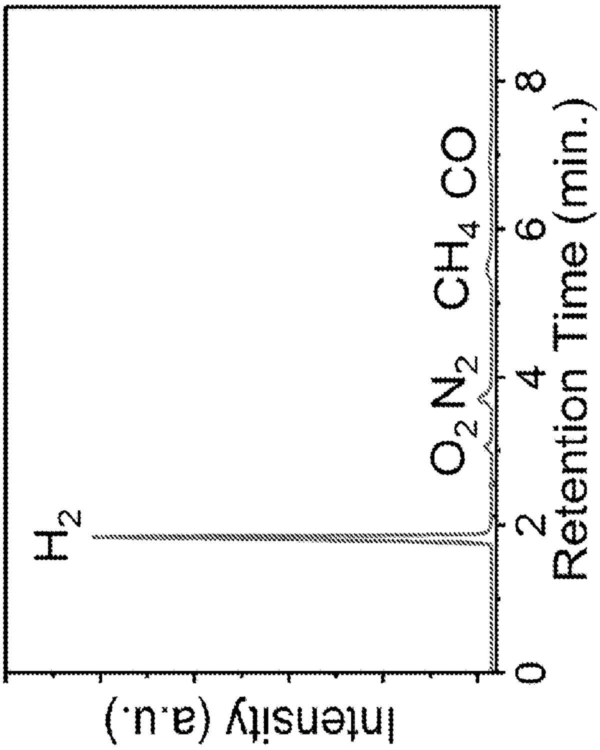

H2
O2 N2
CH4 CO
Intensity (a.u.)
Retention Time (min.)
0
2
4
6
8

FIG. 7B

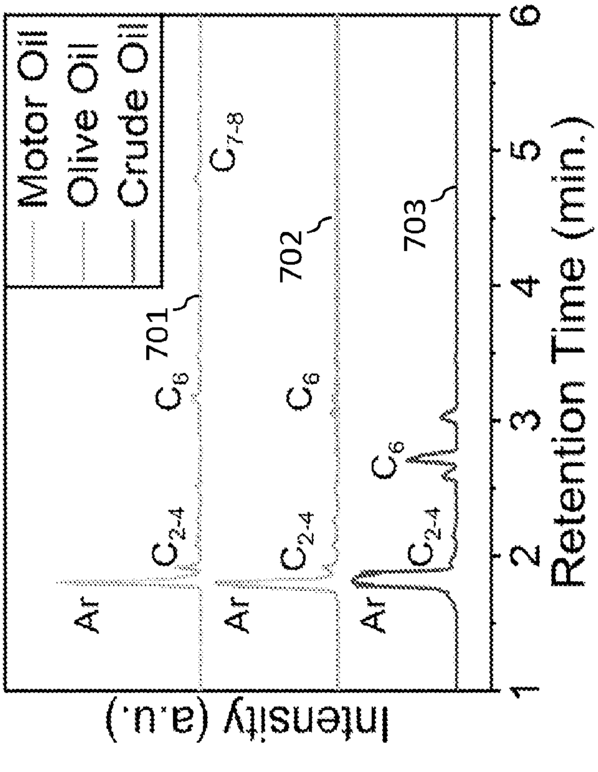

Motor Oil
Olive Oil
Crude Oil
Ar
C2-4
C6
C7-8
701
702
703
Intensity (a.u.)
Retention Time (min.)
1
2
3
4
5
6

FIG. 7A

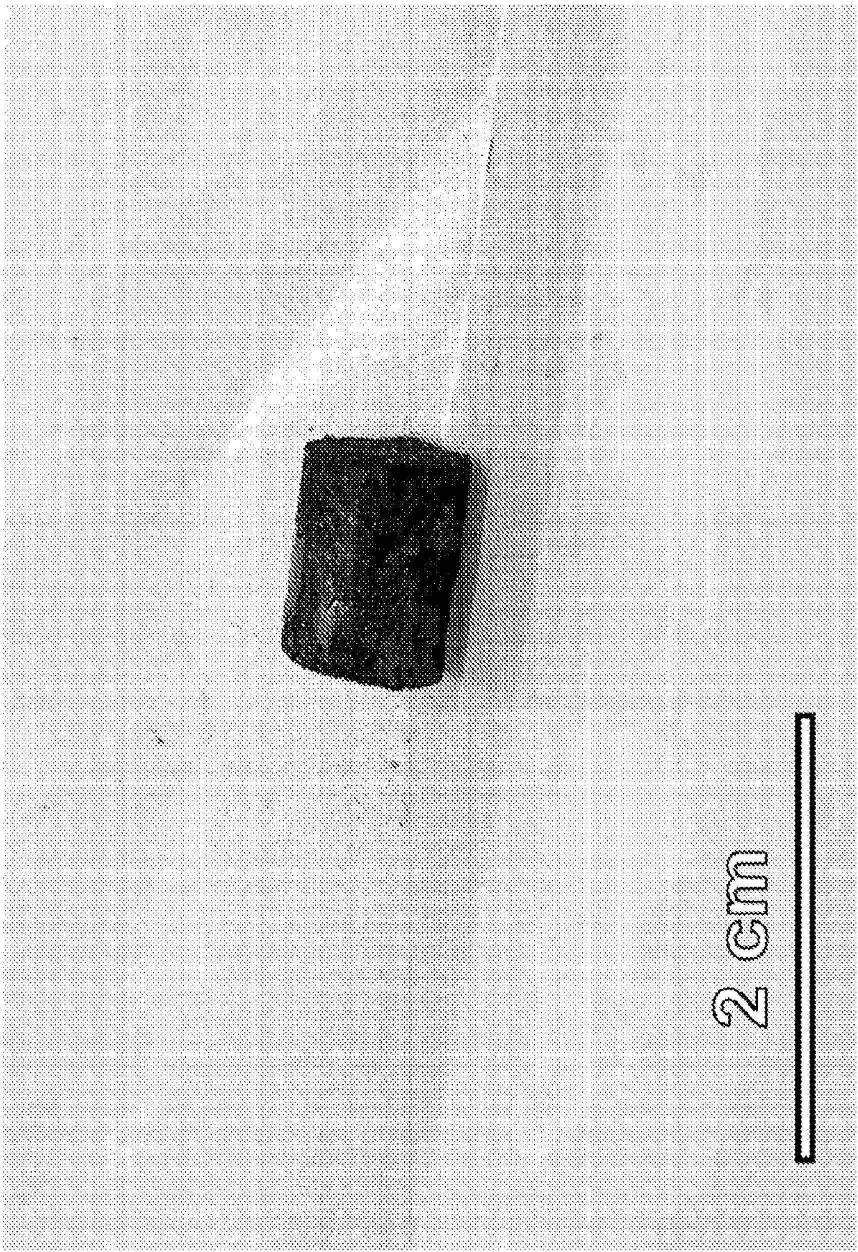


FIG. 8

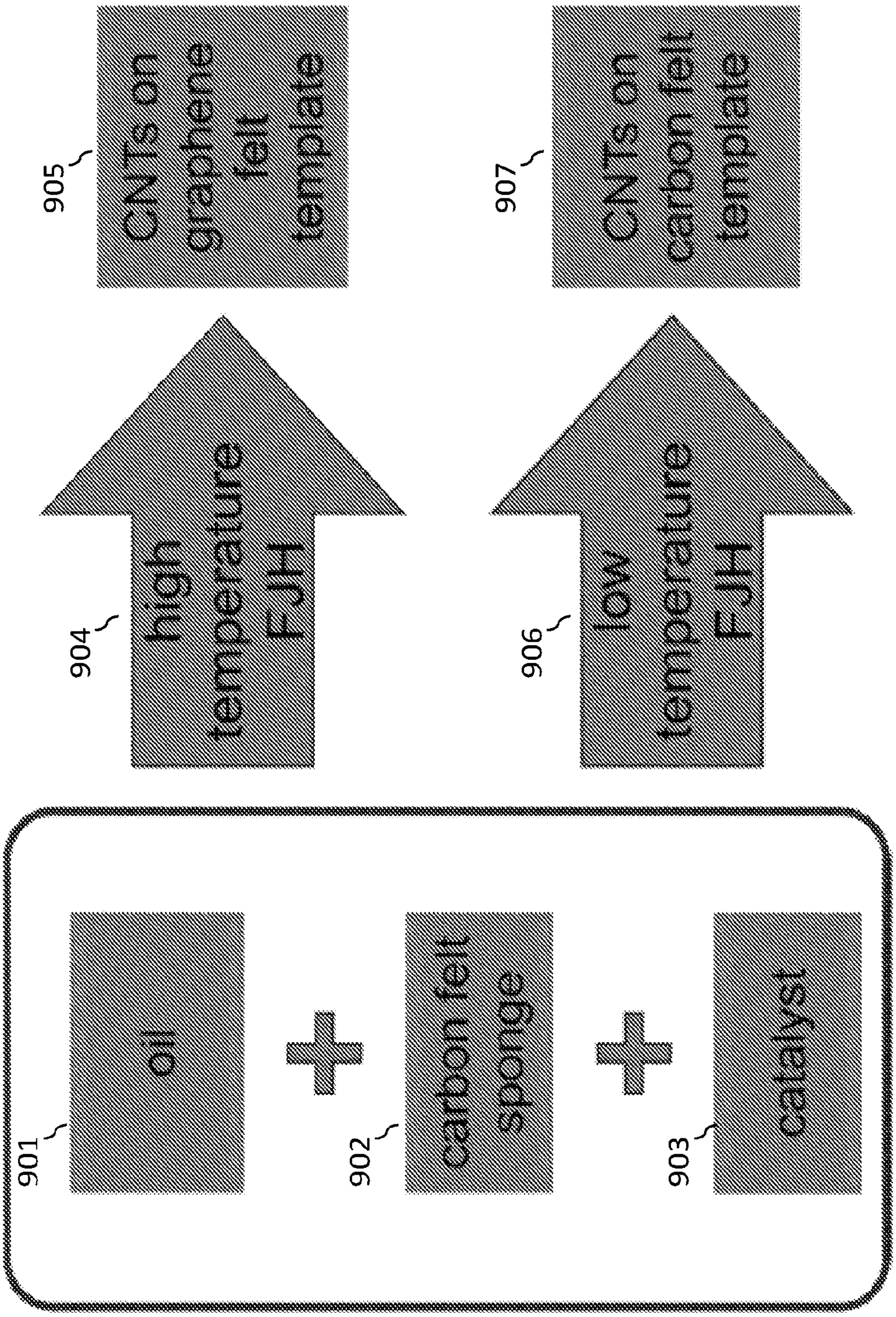

901
oil
902
carbon felt sponge
903
catalyst
904
high temperature FJH
905
CNTs on graphene felt template
906
low temperature FJH
907
CNTs on carbon felt template

FIG. 9

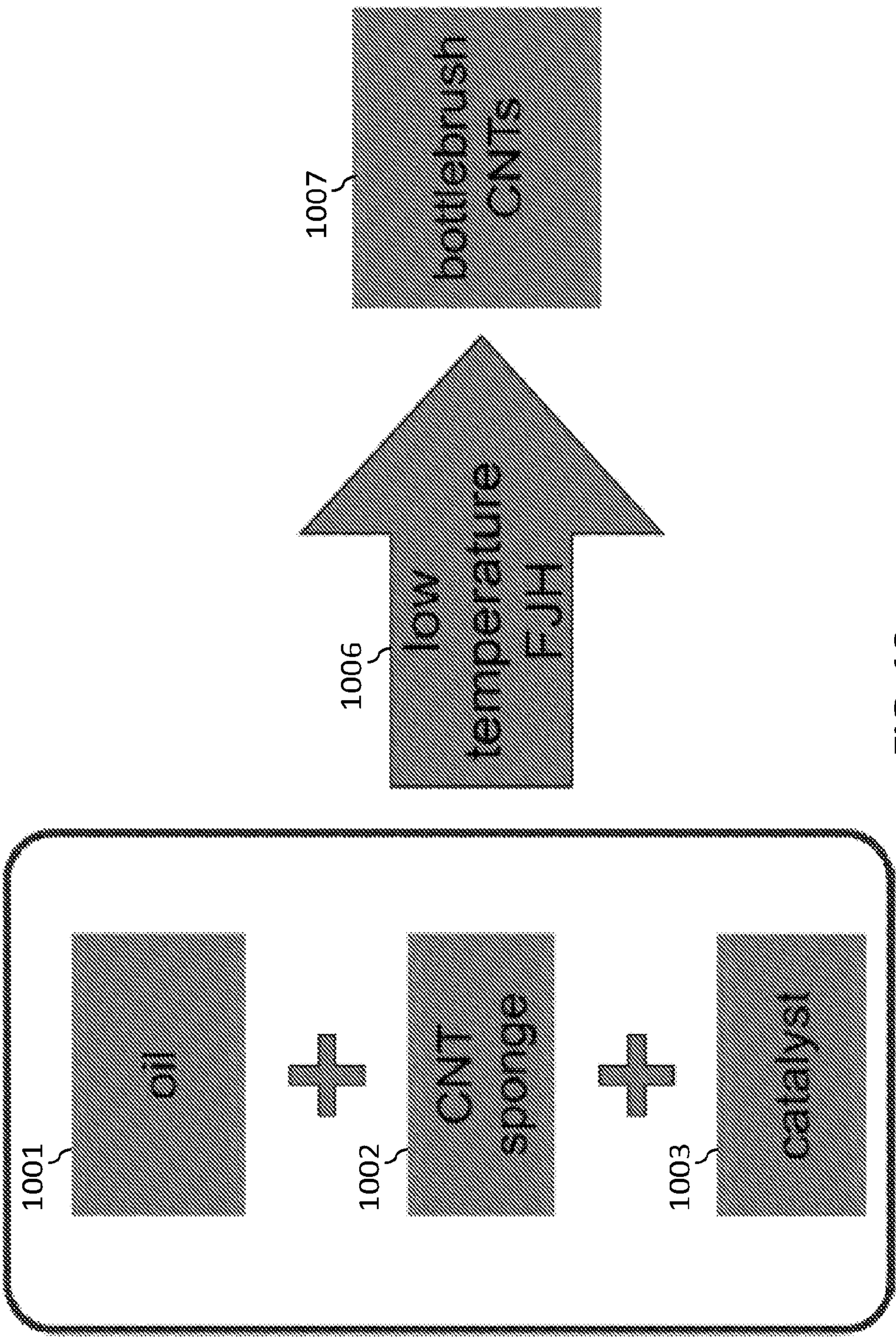

bottlebrush CNTs
1007
low temperature FJH
1006
oil
1001
CNT sponge
1002
catalyst
1003

FIG. 10

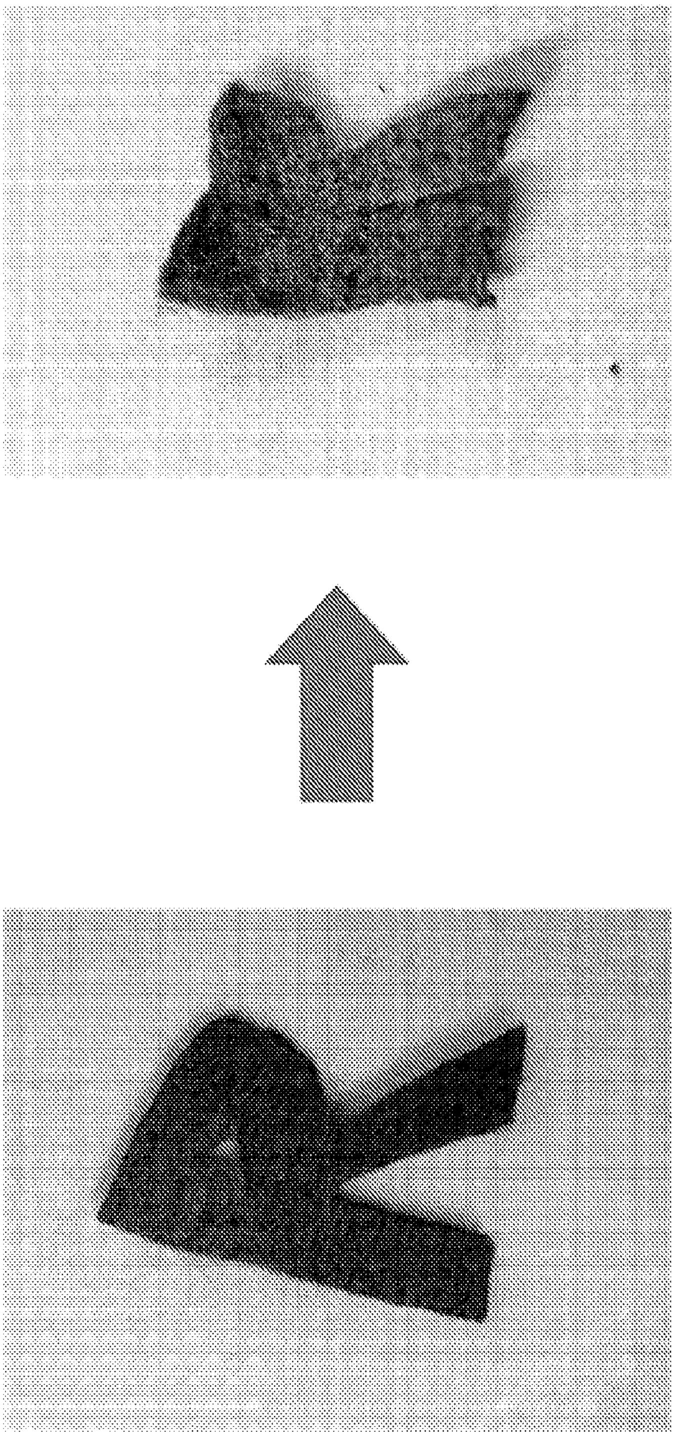


FIG. 11

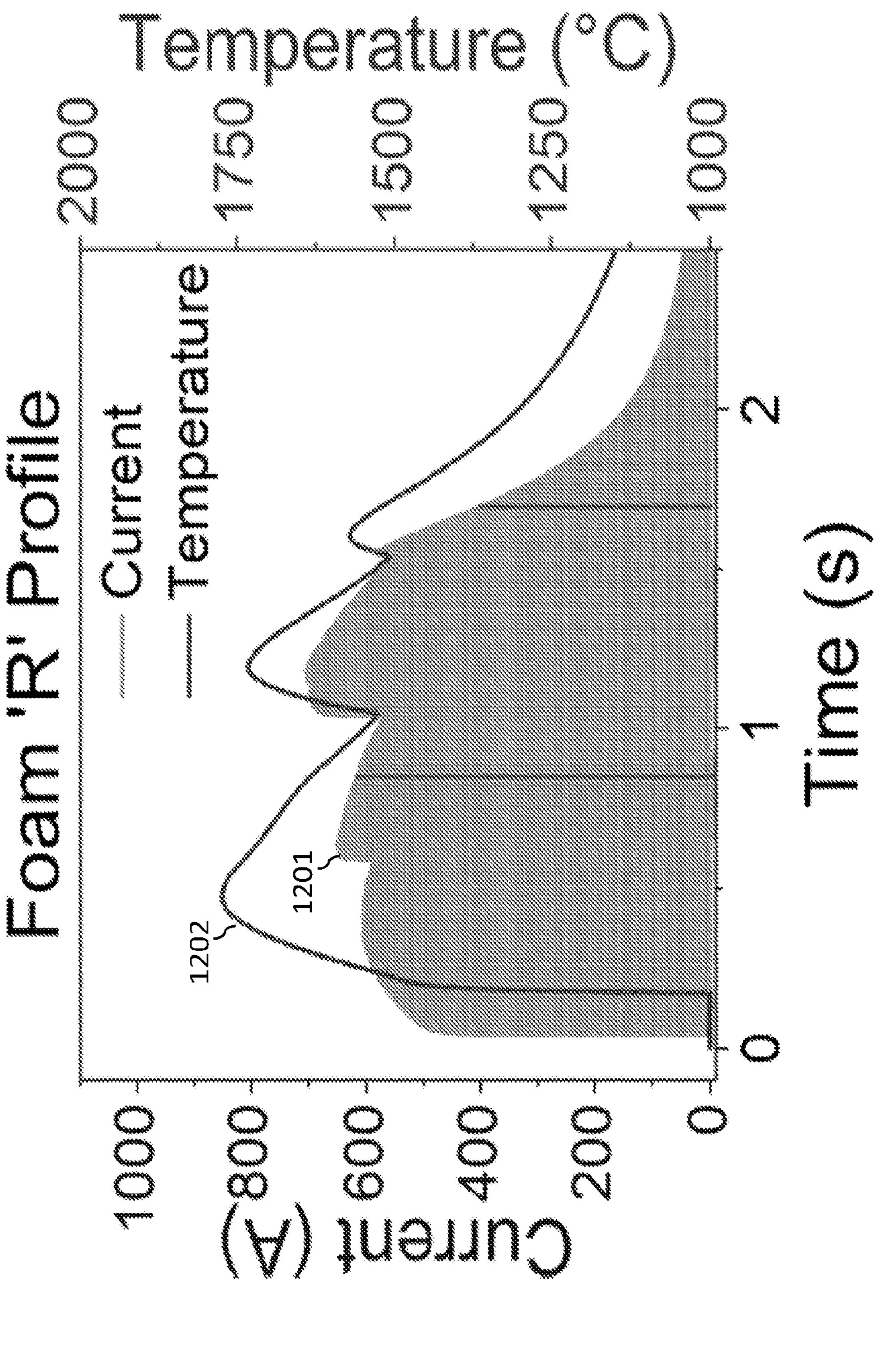

Foam 'R' Profile
Current
Temperature
Temperature (°C)
2000
1750
1500
1250
1000
Current (A)
1000
800
600
400
200
0
Time (s)
0
1
2
1201
1202

FIG. 12

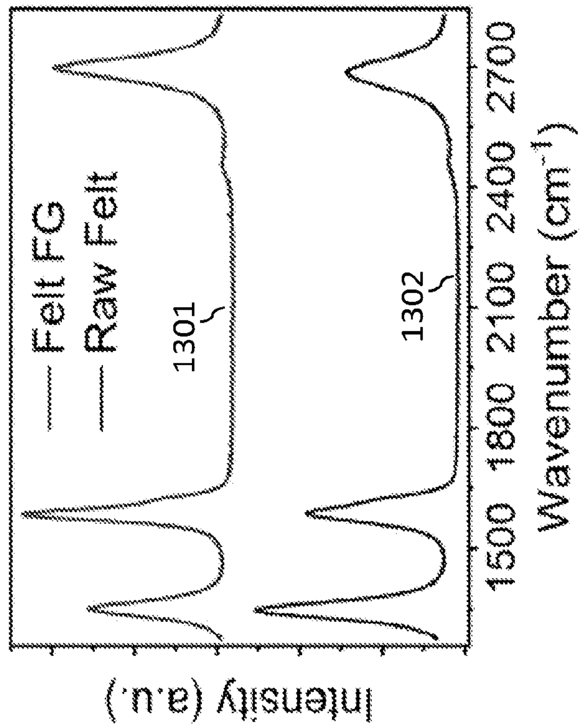


FIG. 13A

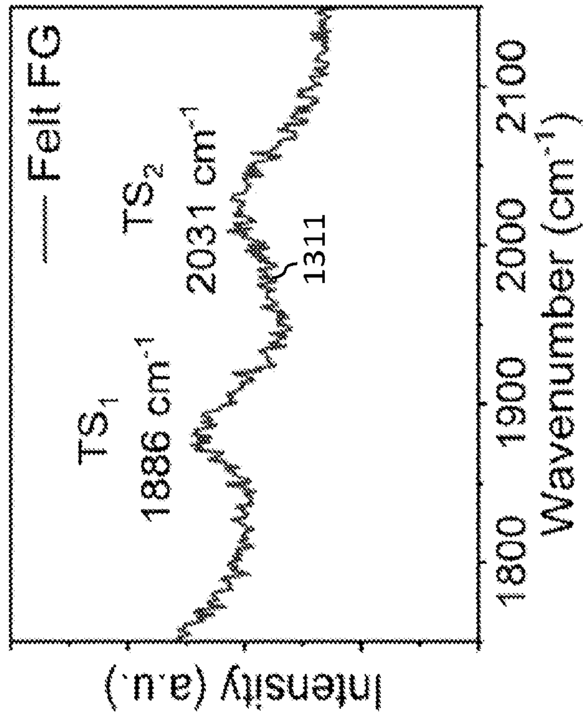


FIG. 13B

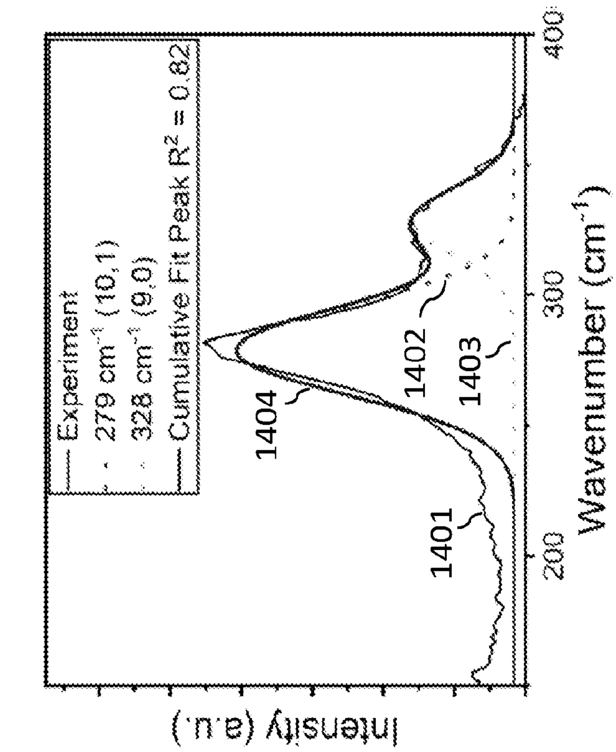


FIG. 14

Raw Carbon Felt

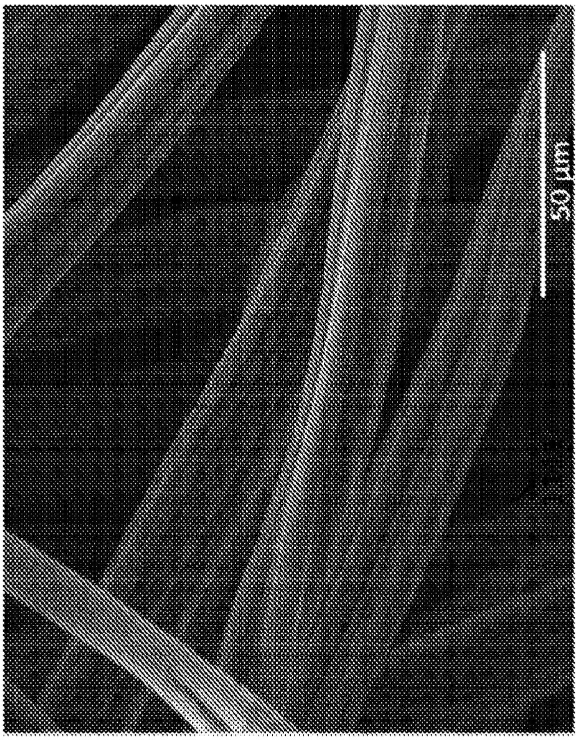


FIG. 15

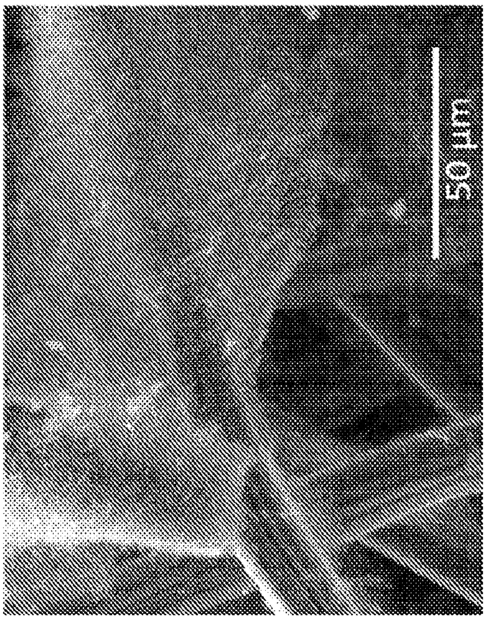


FIG. 16A

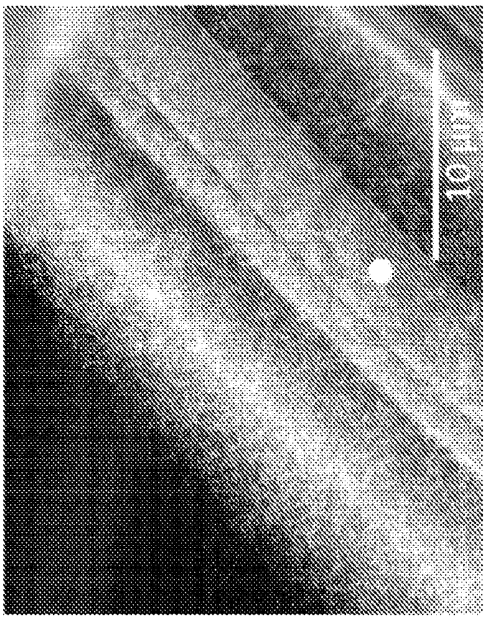


FIG. 16B

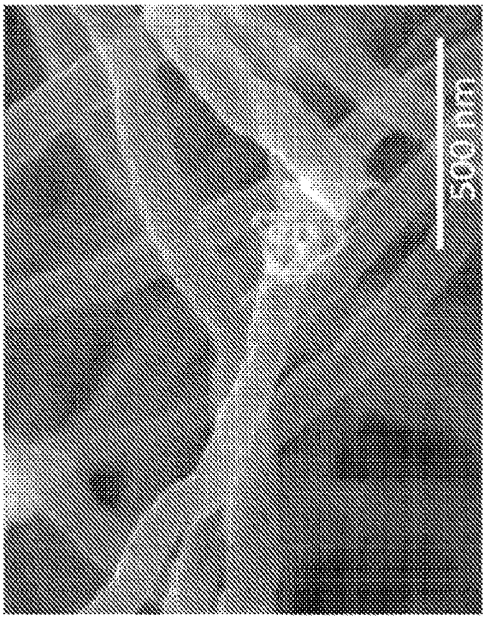


FIG. 16C

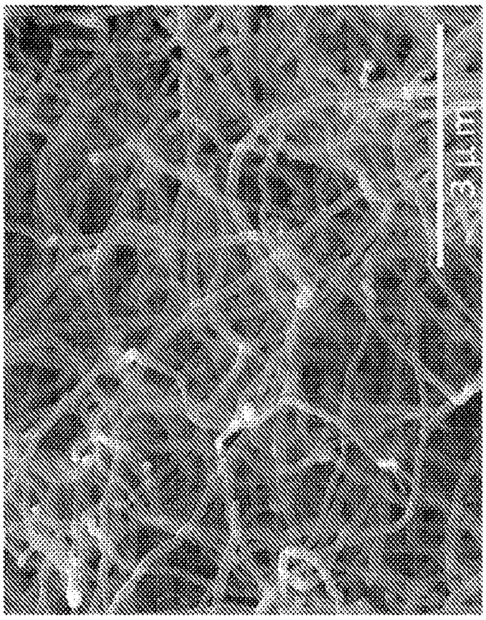


FIG. 16D

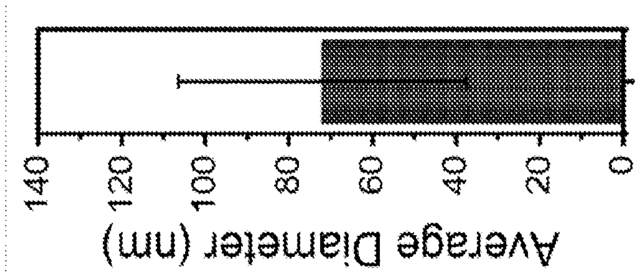


FIG. 17B

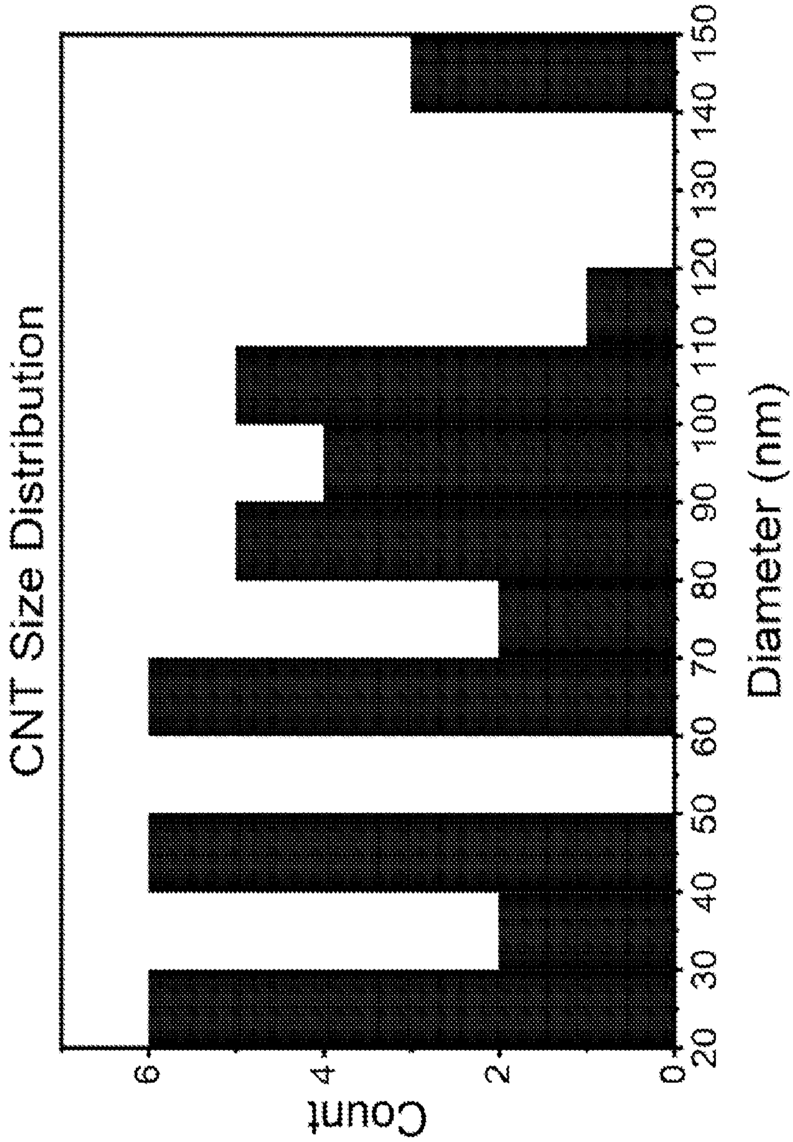


FIG. 17A

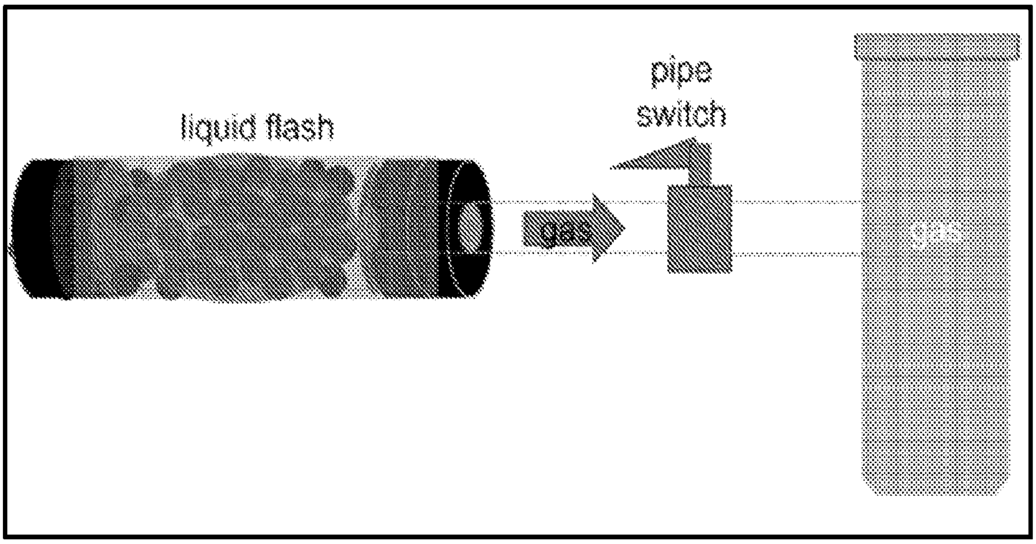


FIG. 18

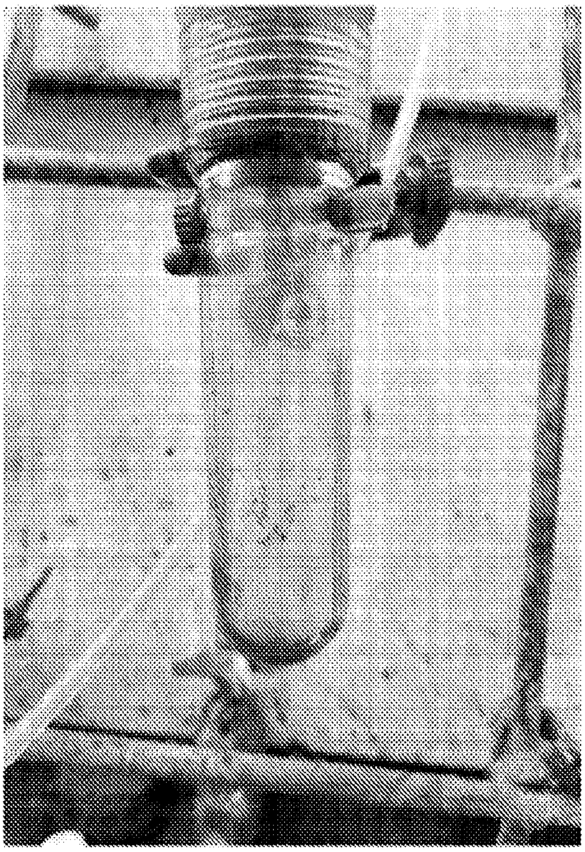


FIG. 19

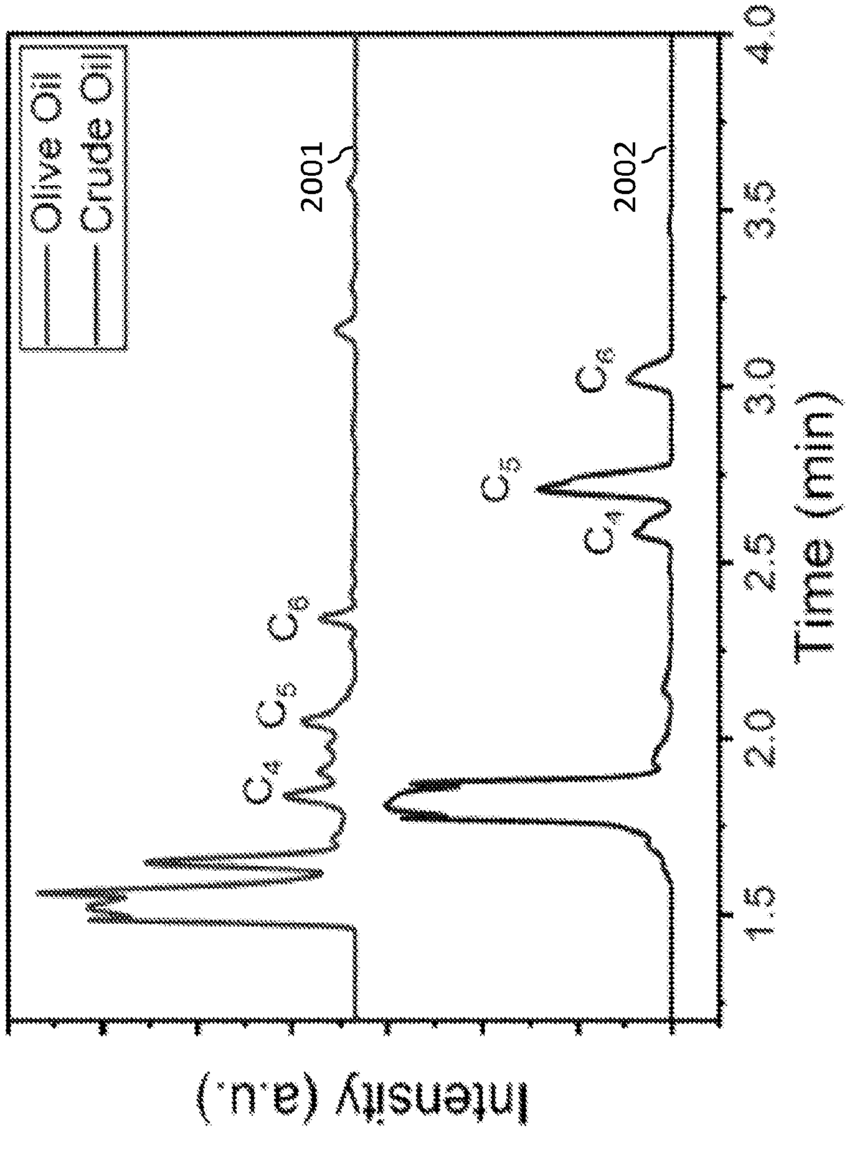

Olive Oil
Crude Oil
2001
2002
C_4
C_5
C_6
C_4
C_5
C_6
1.5
2.0
2.5
3.0
3.5
4.0
Time (min)
Intensity (a.u.)

FIG. 20

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2024/054419

A. CLASSIFICATION OF SUBJECT MATTER

INV. C10G3/00 C10G9/24 C10G1/10 C10B19/00 C10B53/02
C10B53/07 C10B57/06 C10B57/16 C01B3/24 C01B3/26
C01B32/182 C01B32/956 C01B32/05 C01B32/16

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)

C10G C10B C01B

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

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

EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2020/051000 A1 (UNIV RICE WILLIAM M [US]) 12 March 2020 (2020-03-12) claims 1,32,33,126-139,150,151 paragraphs [0234], [0236], [0246] - [0260], [0266] - [0269] figures 1A, 2A -----	1-8, 10-12, 14,15, 17,25, 26,33,34
X	WO 2023/044569 A1 (UNIVERSAL MATTER INC [CA]) 30 March 2023 (2023-03-30) claims 1-3,8,9 paragraphs [0124] - [0129], [0210] -----	1-8, 10-19, 25,26, 33,34
	-/--	

[X] Further documents are listed in the continuation of Box C.

[X] 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

"E" earlier application or patent but published on or after the international filing date

"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)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"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

"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

"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

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
30 January 2025	07/02/2025

Name and mailing address of the ISA/	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Baumlin, Sébastien

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C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2023/192819 A1 (UNIV RICE WILLIAM M [US]) 5 October 2023 (2023-10-05)	1,2, 4-12, 16-22, 25,26, 33,34
Y	claims 1,13-17 paragraphs [0148], [0151] - [0158], [0179] - [0181], [0185], [0192] - [0194] figures 1A, 1B, 1C, 1D -----	23
X	FR 2 831 154 A1 (PHYSIQUES ECP ET CHIMIQUES [FR]) 25 April 2003 (2003-04-25) claim 1 page 15, lines 27-32 table 5 figure 1 -----	1,3,5-8, 10,11, 15-17, 33,34
X	GE JIN ET AL: "Joule-heated graphene-wrapped sponge enables fast clean-up of viscous crude-oil spill", NATURE NANOTECHNOLOGY, 3 April 2017 (2017-04-03), pages 434-440, XP093230840, London ISSN: 1748-3387, DOI: 10.1038/nnano.2017.33 Retrieved from the Internet: URL:http://www.nature.com/articles/nnano.2017.33	1,7, 15-18, 24,27-33
Y	pages 434-435 figures 1, 2a, 2e, 3a, 3b -----	23

1

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/US2024/054419

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		JP 2024535630 A	30-09-2024
		KR 20240067250 A	16-05-2024
		WO 2023044569 A1	30-03-2023
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FR 2831154 A1	25-04-2003	NONE	