are cylindrical nano-structures comprising single or multiple graphitic sheets aligned concentrically or perpendicular to a central axis.

[0016] Secondly, carbon nano-onions (CNOs), which are structures that consist of multiple spherical graphitic sheets that are concentrically layered from a central core, which is typically a catalyst particle or a void; these carbon structures typically range from 50-500 nm in diameter.

[0017] Thirdly, carbon micro-spheres (CMSs), which are hollow globular graphitic structures typically greater than 500 nm in size. They are globular in shape and can be chain-like. Naturally-occurring CMSs are found in meteorites.

[0018] Finally, the parameters of temperature and pressure can be manipulated within the Hazer Process to produce graphene, which is characterised by single-layer or single-digit layer sheets of graphite.

[0019] As with the earlier iteration of the technology described in WO 2016/154666, it will be appreciated that in using a low-grade iron oxide as catalyst (most preferably in the form of Fe_2O_3, Fe_3O_4, hematite ore or goethite ore), the graphite produced, irrespective or morphology, will contain iron-based impurities at a level that requires subsequent purification for its downstream use in many fledgling and established industries.

[0020] Previously, Hazer graphite has been successfully purified to >99.9% w/w using techniques including high temperature thermal purification, and microwave assisted acid digestion. However, these methods can be difficult to scale up and can be expensive.

[0021] It is an object of the present invention to overcome or ameliorate one or more of the disadvantages of the prior art, or at least to provide a useful alternative.

[0022] It is an object of a preferred form of the present invention to provide a process which can purify graphite having a high degree of purity (i.e., greater than 90% and even greater than 99%).

[0023] It is an object of at least one preferred form of the present invention to provide a process which can purify graphite without affecting the morphology or degrading the graphite.

[0024] Although the invention will be described with reference to specific examples, it will be appreciated by those skilled in the art that the invention may be embodied in many other forms.

Summary

[0025] The present disclosure relates generally to a process for increasing the purity of Hazer graphite from 50% to 99.9% (by weight), preferably 80% to 99.7% by electrochemically extracting

the iron impurity. As an additional advantage, it may be possible to recover the iron impurity as a high purity catalyst for the Hazer® Process. The experiments conducted by the inventors demonstrate that the electrochemical purification (ECP) process can purify graphite in both solid and slurry forms. However, the convenience associated with processing Hazer graphite as a slurry is likely to be at the expense of reaction rate and/or power consumption. Results further demonstrate the purification of Hazer graphite at gram scale and the potential for upscaling.

[0026] High-purity graphite finds use in numerous applications including carbon brushes, refractories, batteries, material composites, electrodes, lubricants, coatings, transport, mechanical components, textiles, and household consumer applications.

[0027] Chemical purification of naturally-occurring mined graphite has been used to remove natural impurities from geological formations. However, achieving a highly pure graphite regardless of source (i.e., naturally-occurring or synthetically-produced graphite) via economically-viable means has been problematic. However, some graphite samples (in particular, those produced via the Hazer® Process) have impurities that are heavily encapsulated in the graphitic material and thus are more difficult to remove. Fine milling is typically employed to liberate embedded impurities. However, this is not always practical or possible for nano-sized impurities (such as Hazer® Process graphite). The technique proposed by way of the present disclosure provides a means of addressing this issue.

[0028] The present disclosure provides a process for purifying graphitic material, the process comprising:

> electrochemically treating a crude graphitic material comprising an impurity selected from a metal, metal oxide and combinations thereof;
>
> using a predetermined or regenerative electrolyte;
>
> over a predetermined period;
>
> over a predetermined voltage range;
>
> over a predetermined temperature range;
>
> using a predetermined anode composition;
>
> using a predetermined cathode composition;
>
> thereby removing a portion of the impurity as a result of the electrochemical treatment and providing a purified graphitic material.

[0028a] According to an aspect of the present disclosure, there is provided a process for purifying graphitic material, the process comprising:

electrochemically treating a crude graphitic material comprising an impurity selected from a metal, metal oxide and combinations thereof;

using a predetermined electrolyte;

over a predetermined period;

over a predetermined voltage range;

over a predetermined temperature range;

using a predetermined anode composition;

using a predetermined cathode composition;

thereby removing a portion of the impurity as a result of the electrochemical treatment and providing a purified graphitic material.

[0029] In an example, the process employs a predetermined permeable membrane, which restrains the graphite from contacting the cathode, which can cause a short-circuiting.

[0030] In an example, the impurity is selected from a metal, metal containing impurity, non-metal, non-metal containing impurity, organic, inorganic, and combinations thereof. In a preferred embodiment, the impurity is a non-carbon impurity. In an especially preferred example, the impurity is a metal. Most preferably, the metal is iron or an iron-carbon species such as ferrite, austenite and cementite.

[0031] In another example, the crude graphitic material is compressed prior to use in the process.

[0032] In another example, wherein the process includes no stirring nor agitation.

[0033] In another example, the metal containing impurity is selected from a metal oxide, metal hydroxide, metal nitrate, carbonate, carboxylic acid, salt, and the like.

[0034] In an example, the purified graphitic material has a morphology substantially the same as that of the crude graphitic material. Preferably, the graphitic material has a morphology selected from graphitic fibres (including carbon nano-tubes), carbon nano-onions and carbon micro-spheres.

[0035] In an example, the electrolyte is selected from metals or transition metals with sulfates, sulfites (including bisulfates), phosphates, carbonates, bicarbonates, hydroxides, permanganates, chromates, dichromates, oxalates, formates, acetates, benzoates, halides, chlorites, perchlorites and hypochlorites and the like (e.g., perfluorates, hypobromites, etc.), acids, and the like and mixtures thereof, for instance, sulfate and sulfuric acid. In an especially preferred example, the electrolyte is ammonium sulfate, iron sulfate or a mixture thereof. In another example, the electrolyte is sulfuric acid or nitric acid. Preferably, the electrolyte is iron sulfate or ammonium sulfate. Those skilled in the art will appreciate that it is possible to use any suitable salt provided that the salt or salts selected form a soluble compound and/or complex with iron and or metal impurities. In further example, the electrolyte is selected from sulfides, phosphides, phenolates, superoxides, peroxides, oxides, silicates, sulfones, thiocyanates, thiosulfates, selenates, triiodides, azides, cyanides, cyanates, borates, fulminates, arsenates, vanadates, antimonates and the like.

[0036] In an example, the iron impurity precipitates as iron dendrites on the cathode. Typically, the iron dendrites are harvested and applied industrially. In some examples, the iron dendrites are used as catalyst in the thermocatalytic decomposition of methane to hydrogen

and iron-contaminated graphitic material. In other examples, the iron can be in the form of a sludge. In certain examples, the sludge can be recovered and refined and/or tuned to catalyst, for example low – or high-performing catalyst depending on the post-processing methodology. In a preferred example, the sludge, once refined to produce an iron containing catalyst, can be used in the Hazer process to produce hydrogen and graphite from hydrocarbons, preferably methane.

[0037] In an example, the iron impurity remains in solution. In an example, the iron impurity is precipitated from solution to be used as catalyst in the thermocatalytic decomposition of methane to hydrogen and iron-contaminated graphitic material such as the Hazer® process.

[0038] The precipitated iron can have a specific shape, size and purity or composition (pure iron or a complex). The precipitation can be achieved as part of the cell or as a separate process.

[0039] In an example, the iron impurity is precipitated from solution to be used as catalyst in the thermocatalytic decomposition of methane to hydrogen and iron-contaminated graphitic material. In an example, the iron impurity, once precipitated, is dried, crushed, and / or filtered.

[0040] In an example, the iron-contaminated graphitic material is subsequently purified by a process as defined herein.

[0041] In an example, the process is performed on a continuous, substantially continuous or batch basis.

[0042] In certain examples, the process is performed on a batch basis with each batch comprising about 1 g to 3 kg of graphitic material. For example, each batch comprises about 1 g to about 10 g, or about 10 g to about 20 g, or about 20 g to about 30 g, or about 30 g to about 40 g, or about 40 g to about 50 g, or about 50 g to about 60 g. or about 60 g to about 70 g, or about 70 g to about 80 g, or about 80 g to about 90 g, or about 90 g to about 100 g, or about 100 g to about 200 g, or about 200 g to about 300 g, or about 300 g to about 400 g, or about 400 g to about 500 g, or about 500 g to about 600 g, or about 600 g to about 700 g, or about 700 g to about 800 g, or about 800 g to about 900 g, or about 900 g to about 1 kg, or about 1 kg to about 2 kg, or about 2 kg to about 3 kg of graphite material. In an example, the process does not lose efficiency or loses minimal efficiency with the scale-up of the amount of graphitic material in each batch.

[0043] In an example, the voltage range is between about 1 V and 300 V. Preferably, the voltage range is between about 5 and 300V. In a specific example, the voltage is about 20 V. It was found that a voltage of 20 V increases purification over 24 hours and increases reaction kinetics, as compared to other voltage ranges. However, it may not increase the final purity of the

graphitic material.

[0044] In an example, the current is constant in the process. In other example, the current is varied in the process. In certain examples, the current may be increased to increase the rate of reaction in the process.

[0045] In an example, the period is between about 30 minutes and about 2 weeks. In another embodiment, the period is between about 2 h and about 96 h. Preferably, the period is between about 24 h and about 48 h.

[0046] In an example, the temperature range is between about 5 ^{o}C and about 100 ^{o}C. In an example, the temperature range is between about 20 ^{o}C and about 80 ^{o}C. In an embodiment, the temperature range is between about 40 ^{o}C and about 60 ^{o}C.

[0047] In an example, the anode comprises one or more structures. Preferably, the structures comprise rods, plates, filaments and the like. In an example, the structures are crystalline or non-crystalline. In an example, the structures are graphitic. In an example, the structures comprise platinum or are titanium coated with platinum, preferably are platinum. In an example, the anode comprises graphite, lead, lead alloys, platinum, titanium coated with platinum and combinations thereof. In a particularly preferred example, the structures are titanium coated with platinum.

[0048] In an example, the cathode comprises graphite, lead, lead alloys, platinum, titanium coated with platinum and combinations thereof. In an example, the cathode comprises platinum, titanium coated with platinum and graphitic electrodes, various grades of stainless steel, ferrous alloys, other transition metal alloys and the like. In a preferred example, the cathode comprises titanium coated with platinum.

[0049] In an example, the process further comprises the use of a permeable membrane covering at least a portion of the anode, cathode, or both. Preferably, the permeable membrane is a neutrally-charged permeable membrane, an anion exchange membrane, or a cation exchange membrane. More preferably, the permeable membrane is selected from asbestos cloth, cellulose, glass cloth, filter cloth, glass cloth impregnated with silica gel, porous sintered stainless steel, vinyl chloride acrylonitrile, polysulfone, polyethersulfone (PES), polycarbonate, polytetrafluoroethylene, polyethylene terephthalate (PET) and combinations thereof. Alternatively, the membrane may comprise sintered metal and non-metal materials such as ceramics.

[0050] In an example, the permeable membrane has a molecular weight cut-off (MWCO) of

less than about 1 million Da. Preferably, the permeable membrane has a molecular weight cut-off (MWCO) of between about 10 kDa, and about 0.5 kDa. More preferably, the permeable membrane has a molecular weight cut-off (MWCO) of about 3.5 kDa. In one example, the permeable membrane has an air permeability of between 0.1 and 100 L/min/dm^2 at 200 Pa. Preferably, the permeable membrane has an air permeability of between 1 and 50 L/min/dm^2 at 200 Pa. More preferably, the permeable membrane has an air permeability of between 2 and 30 L/min/dm^2 at 200 Pa.

[0051] In an example, the purified graphitic material has a purity of greater than about 95% w/w. Preferably, the purified graphitic material has a purity of greater than about 99% w/w. More preferably, the purified graphitic material has a purity of greater than about 99.5% w/w. Most preferably, the purified graphitic material has a purity of greater than about 99.9% w/w.

[0052] In an example, the purified graphitic material is used as the crude graphitic material in the process to provide iterative purification.

[0053] The present disclosure provides a purified graphitic material, when purified by a process as defined herein.

[0053a] According to another aspect of the present disclosure, there is provided a purified graphitic material, obtained or obtainable from a process as defined herein.

[0054] The present Inventors have surprisingly found a novel, facile approach for the purification of graphite at relatively low cost. The electrochemical processes employed (an electrolytic cell or electrorefining), due to their simple operation give rise to a surprisingly-efficient relatively low-cost process for the purification of graphite.

[0055] In some examples, the impurity undergoes a redox reaction during electrochemical treatment which removes a portion of the impurity from the graphite. In some examples, the impurity that undergoes a redox reaction during electrochemical treatment forms a salt such as a metal salt. In some examples, the impurity that undergoes a redox reaction during electrochemical treatment forms a water-soluble salt. In some examples, the impurity which undergoes a redox reaction during electrochemical treatment forms a water-insoluble salt.

[0056] In some examples, the impurity is iron. The iron can be synthetic (e.g., from Fe_3O_4) or naturally occurring (e.g., from hematite) or elemental iron or iron carbide or the like. In some examples, the impurity that undergoes a redox reaction during electrochemical

treatment forms a cationic species. In some examples, the impurity that undergoes a redox reaction during electrochemical treatment forms an iron cationic species such as Fe^{3+}.

[0057] In some examples, the electrochemical treatment does not substantially affect the

morphology of, or damage, the graphite. Advantageously, purification without damaging or affecting the morphology of the treated graphite can increase the yield and value of the purified graphite.

[0058] In some examples, the process comprises the use of a permeable membrane between a cathode and an anode during electrochemical treatment. The membrane primarily serves to prevent the graphite from contacting the cathode and short-circuiting the electrochemical cell.

[0059] In some examples, the salt formed by the redox reaction further undergoes a salt metathesis reaction. In certain examples, the salt metathesis reaction forms an insoluble salt. In some examples, the insoluble salt formed by the salt metathesis reaction is iron hydroxide or an iron complex such as jarosite. Advantageously, the insoluble salt, such as iron hydroxide, can be a valuable by-product formed during the purification process of the present disclosure. For example, iron hydroxide can be used as a catalyst, such as for the decomposition of methane to form graphite and hydrogen gas, most conveniently via the Hazer® Process. As would be appreciated by a skilled addressee, a variety of insoluble iron species can be used as a catalyst for the Hazer® process, either directly or indirectly.

[0060] In some examples, the electrolyte is a sulfate salt. In some examples, the electrolyte is selected from the group consisting of an ammonium sulfate, iron sulfate, sulfuric acid and combinations thereof. Advantageously, when iron sulfate is used as the electrolyte, elemental iron forms on the cathode during the electrochemical treatment of the impure graphite. The elemental iron can be in the form of dendrites and is formed instead of insoluble iron hydroxide. However, those skilled in the art will understand that cell can be tuned so the depositing metal does not form dendrites and instead forms a uniform metallic coating around the cathode (which may be preferable); dendrites are not generally preferred as they may cause a short circuit. Similarly, elemental iron can be a valuable by-product formed during the purification process of the present disclosure. For example, iron can also be used as a catalyst, such as for decomposition of methane to form graphite and hydrogen gas, again, most conveniently via the Hazer® Process.

[0061] In some examples, iron in solution is preferred, or elemental iron is precipitated as an insoluble species instead of depositing the elemental iron onto the cathode.

[0062] In some examples, the impurity is naturally-derived, for example, natural impurities from geological formations such as mined graphite. In some examples, the impurity is synthetically derived, for example the impurity is introduced as a result of synthetically producing the graphite. For example, the process described in WO 2016/000115, as related above. This

publication, along with WO 2017/031529 is fully incorporated herein by reference in their respective entireties.

[0063] In some examples, the purified graphite has a purity of greater than 90% (by weight), preferably greater than 95%, more preferably greater than 98% and yet more preferably greater than 99%. In some examples, the purified graphite has a purity greater than about 80, 81, 82, 83, 84, 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 96, 97, 98 or 99%. In some examples, the purified graphite has a purity greater than about 98.1, 98.2, 98.3, 98.4, 98.5, 98.6, 98.7, 98.8, 98.9, 99.0, 99.1, 99.2, 99.3, 99.4, 99.5, 99.6, 99.7, 99.8 or 99.9%. In some examples, the purified graphite has a purity greater than about 99.95%.

[0064] According to another form, the present disclosure provides a process for purifying graphite comprising: electrochemically treating a graphite containing an impurity selected from an iron, iron oxide and combinations thereof in the presence of an electrolyte comprising a sulfate salt; thereby removing a portion of the impurity as a result of the electrochemical treatment and providing a purified graphite.

[0065] The present disclosure provides a purified graphite comprising an impurity selected from a metal, metal oxide and combinations thereof, wherein the purified graphite has a concentration of impurity less than 20% w/w.

[0066] In some examples, the purified graphite has a concentration of impurity less than 15% w/w, preferably less than 10%, preferably less than 5%, more preferably less than 3%, more preferably less than 2%, yet more preferably less than 1%. In some examples, the purified graphite has a concentration of impurity less than 20%, 19%, 18%, 17%, 16%, 15%, 14%, 13%, 12%, 11%, 10%, 9%, 8%, 7%, 6%, 5%, 4%, 3%, 2% or 1% w/w. In some examples, the purified graphite has a concentration of impurity less than 0.9, 0.8, 0.7, 0.6, 0.5, 0.4, 0.3, 0.2, 0.1 or 0.05% w/w.

[0067] The present disclosure provides a negative electrode material comprising a coating on a substrate, the coating comprising: MnO_2 (electrolytic manganese dioxide (EMD)), a carbon conductive additive in the form of purified graphitic material as defined herein, and a binder.

[0067a] According to another aspect of the present disclosure, there is provided a negative electrode comprising a coating on a substrate, the coating comprising: MnO_2 (electrolytic manganese dioxide (EMD)), a carbon conductive additive in the form of purified graphitic material as defined herein, and a binder.

[0068] In some examples, the EMD is one or more of α-, β-, γ-, δ-, or λ-MnO_2.

[0069] In some examples, the EMD is comprised of substantially α-, β-, γ-, δ-, or λ-MnO_2. In some examples, the EMD is γ-MnO_2.

[0070] In some examples, the EMD, the carbon conductive additive and the binder are mixed together in a weight ratio of 4-9:2:1.

[0071] In some examples, the EMD, the carbon conductive additive and the binder are mixed together in a weight ratio of 7:0.1-3:1.

[0072] In some examples, the EMD, the carbon conductive additive and the binder are mixed together in a weight ratio of 7:2:0.1-3.

[0073] In some examples, the EMD, the carbon conductive additive and the binder are mixed together in a weight ratio of 4-9:0.1-3:0.1-3.

[0074] In one example, the EMD, the carbon conductive additive and the binder are mixed together in a weight ratio of 7:2:1

[0075] In some examples, the binder comprises a fluoropolymer selected from the group consisting of polyvinylidene fluoride (PVDF), polytetrafluoroethylene (PTFE), fluorinated ethylene propylene (FEP), perfluoroalkoxyl polymer (PFA) and polyvinyl fluoride (PVF).

[0076] In some examples, the binder is selected from the group consisting of carboxymethyl cellulose (CMC), sodium alginate, starch, styrene-butadiene rubber (SBR), xanthan gum, polyvinyl chloride (PVC), polyacrylic acid (PAA), polyvinyl alcohol (PVA), polyethylene glycol (PEG), and polyamide imide (PAI).

[0077] In one example, the binder comprises polyvinylidene fluoride (PVDF).

[0078] In some examples, the substrate comprises a metal foil.

[0079] In some examples, the metal foil is made of a conductive metal.

[0080] In some examples, the conductive metal is copper, zinc, aluminium, iron or any mixture thereof.

[0081] In some examples, the coating at least partially surrounds the substrate.

[0082] In some examples, the coating surrounds the substrate.

[0083] In some examples, the coating has a thickness in the range of about 1 micron to about 25 microns.

[0084] In some examples, the coating has a thickness of about 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24 or about 25 microns.

[0085] In some examples the coating has a thickness in the range of about 5 microns to about

20 microns.

[0086] In some examples the coating has a thickness in the range of about 7 microns to about 15 microns.

[0087] In one example, the coating has a thickness of about 10 microns.

[0088] In some examples, the negative electrode has an electrical conductivity that falls within a range of about 70 S m^{-1} to about 100 S m^{-1}.

[0089] In some examples, the negative electrode has an electrical conductivity of about 70, 71, 72, 73, 74, 75, 76, 77, 78, 79, 80, 81, 82, 83, 84, 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 96, 97, 98, 99 or about 100 S m^{-1}.

[0090] In some examples, the negative electrode has an electrical conductivity that falls within a range of about 80 S m^{-1} to about 95 S m^{-1}.

[0091] In one example, the negative electrode has an electrical conductivity of about 90 S m^{-1}.

[0092] The present disclosure provides a battery comprising:

a positive electrode;

a negative electrode; and

an electrolyte in contact with the positive electrode and the negative electrode,

wherein the negative electrode comprises a coating on a substrate, and wherein the coating comprises MnO_2 (electrolytic manganese dioxide (EMD)), a carbon conductive additive in the form of purified graphitic material as defined herein, and a binder.

[0092a] According to another aspect of the present disclosure, there is provided a battery comprising:

a positive electrode;

a negative electrode as defined herein; and

an electrolyte in contact with the positive electrode and the negative electrode.

[0093] In some examples, the EMD is one or more of α-, β-, γ-, δ-, or λ-MnO_2.

[0094] In some examples, the EMD is comprised of substantially α-, β-, γ-, δ-, or

λ-MnO_2.

[0095] In some examples, the EMD is γ-MnO_2.

[0096] In some examples, the EMD, the carbon conductive additive and the binder are mixed together in a weight ratio of 4-9:2:1.

[0097] In some examples, the EMD, the carbon conductive additive and the binder are mixed together in a weight ratio of 7:0.1-3:1.

[0098] In some examples, the EMD, the carbon conductive additive and the binder are mixed together in a weight ratio of 7:2:0.1-3.

[0099] In some examples, the EMD, the carbon conductive additive and the binder are mixed together in a weight ratio of 4-9:0.1-3:0.1-3.

[00100] In one example, the EMD, the carbon conductive additive and the binder are mixed together in a weight ratio of 7:2:1

[00101] In some examples, the binder comprises a fluoropolymer selected from the group consisting of polyvinylidene fluoride (PVDF), polytetrafluoroethylene (PTFE), fluorinated ethylene propylene (FEP), perfluoroalkoxyl polymer (PFA) and polyvinyl fluoride (PVF).

[00102] In some examples, the binder is selected from the group consisting of carboxymethyl cellulose (CMC), sodium alginate, starch, styrene-butadiene rubber (SBR), xanthan gum, polyvinyl chloride (PVC), polyacrylic acid (PAA), polyvinyl alcohol (PVA), polyethylene glycol (PEG), and polyamide imide (PAI).

[00103] In one example, the binder comprises polyvinylidene fluoride (PVDF).

[00104] In some examples, the substrate comprises a metal foil.

[00105] In some examples, the metal foil is made of a conductive metal.

[00106] In some examples, the conductive metal is copper, zinc, aluminium, iron or any mixture thereof.

[00107] In some examples, the coating at least partially surrounds the substrate.

[00108] In some examples, the coating surrounds the substrate.

[00109] In some examples, the coating has a thickness in the range of about 1 micron to about 25 microns.

[00110] In some examples, the coating has a thickness of about 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24 or about 25 microns.

[00111] In some examples the coating has a thickness in the range of about 5 microns to about 20 microns.

[00112] In some examples the coating has a thickness in the range of about 7 microns to about 15 microns.

[00113] In one example, the coating has a thickness of about 10 microns.

[00114] In some examples, the negative electrode has an electrical conductivity that falls within a range of about 70 S m^{-1} to about 100 S m^{-1}.

[00115] In some examples, the negative electrode has an electrical conductivity of about 70, 71, 72, 73, 74, 75, 76, 77, 78, 79, 80, 81, 82, 83, 84, 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 96, 97, 98, 99 or about 100 S m^{-1}.

[00116] In some examples, the negative electrode has an electrical conductivity that falls within a range of about 80 S m^{-1} to about 95 S m^{-1}.

[00117] In one example, the negative electrode has an electrical conductivity of about 90 S m^{-1}.

[00118] In some examples, the electrolyte is an aqueous electrolyte.

[00119] In some examples, the electrolyte is an aqueous electrolyte present in a concentration that falls within the range of about 0.01 M to about 10 M.

[00120] In some examples, the electrolyte is an aqueous electrolyte present in a concentration of about 0.01, 0.05, 0.1, 0.15, 0.2, 0.25, 0.3, 0.35, 0.4, 0.45, 0.5, 0.55, 0.6, 0.65, 0.7, 0.75, 0.8, 0.95, 1.0, 1.5, 2.0, 2.5, 3.0, 3.5, 4.0, 4.5, 5.0, 5.5, 6.0, 6.5, 7.0, 7.5, 8.0, 8.5, 9.0, 9.5 or about 10 M.

[00121] In some examples, the electrolyte is an aqueous electrolyte present in a concentration that falls within the range of about 0.01 M to about 5 M.

[00122] In some examples, the electrolyte is an aqueous electrolyte present in a concentration that falls within the range of about 0.01 M to about 2 M.

[00123] In some examples, the electrolyte is an aqueous electrolyte selected from the group consisting of a sulfate salt, nitrate salt, chloride salt and combinations thereof.

[00124] In some examples, the electrolyte is an aqueous electrolyte selected from the group consisting of an ammonium sulfate, sodium sulfate, magnesium sulfate, iron sulfate, copper sulfate, zinc sulfate ($ZnSO_4$), cadmium nitrate, cobalt nitrate, sodium nitrate, sodium chloride, nickel chloride, potassium chloride, ammonium chloride, calcium chloride, sulfuric acid and combinations thereof.

[00125] In some examples, the electrolyte is zinc sulfate ($ZnSO_4$), zinc chloride ($ZnCl_2$), ammonium chloride (NH_4Cl), and zinc trifluoromethyl sulfonate ($Zn(CF_3SO_3)_2$) or mixtures thereof.

[00126] In some examples, the electrolyte is an ionic liquid comprising a cation selected from the group consisting of 1-alkyl-3-methyl-imidazolium, *N*-alkyl-pyridinium, tetraalkyl-

ammonium, tetralkyl-phosphonium and combinations thereof.

[00127] In some examples, the alkyl group is selected from the group consisting of C_2-C_{12} alkyl.

[00128] In some examples, the ionic liquid comprises a cation selected from the group consisting of 1-ethyl-3-methyl-1*H*-imidazolium, 1-butyl-3-methyl-1*H*-imidazolium, 1-butylpyridinium and combinations thereof.

[00129] In some examples, the electrolyte is $ZnSO_4$.

[00130] In some examples, the $ZnSO_4$ electrolyte is present in a concentration that falls within the range of about 0.01 M to about 10 M.

[00131] In one example, the $ZnSO_4$ electrolyte is present in a concentration of 1.0 M.

[00132] In some examples, the positive electrode is a zinc metal electrode.

[00133] In some examples, the negative electrode has a specific discharge capacity that falls within a range of between about 50 mAh g^{-1} to about 200 mAh g^{-1}, when measured between 1.5 V and 0.7 V under a current density of 0.05 A g^{-1}.

[00134] In some examples, the negative electrode has a specific discharge capacity that falls within a range of between about 70 mAh g^{-1} to about 120 mAh g^{-1}, when measured between 1.5 V and 0.7 V under a current density of 0.05 A g^{-1}.

[00135] In one example, the negative electrode has a specific discharge capacity of at least about 109 mAh g^{-1}, when measured between 1.5 V and 0.7 V under a current density of 0.05 A g^{-1}.

Definitions

[00136] In describing and defining the present invention, the following terminology will be used in accordance with the definitions set out below. It is also to be understood that the terminology used herein is for the purpose of describing particular embodiments of the invention only and is not intended to be limiting. Unless defined otherwise, all technical and scientific terms used herein have the same meaning as commonly understood by one having ordinary skill in the art to which the invention pertains.

[00137] The terms “graphite” and “graphitic material” are generally considered synonymous for the purposes of the present invention. This definition includes carbon materials that are crystalline (short and long-range crystallinity), and most preferably includes the various

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:-

1. A process for purifying graphitic material, the process comprising:

electrochemically treating a crude graphitic material comprising an impurity selected from a metal, metal oxide and combinations thereof;

using a predetermined electrolyte;

over a predetermined period;

over a predetermined voltage range;

over a predetermined temperature range;

using a predetermined anode composition;

using a predetermined cathode composition;

thereby removing a portion of the impurity as a result of the electrochemical treatment and providing a purified graphitic material.

2. A process according to claim 1, further comprising a predetermined semipermeable membrane to protect the cathode from short-circuiting upon contacting the graphitic material.

3. A process according to claim 1 or claim 2, wherein the purified graphitic material has a morphology substantially the same as that of the crude graphitic material, wherein the graphitic material has a morphology selected from graphitic fibres (including carbon nano-tubes), carbon nano-onions, carbon micro-spheres and graphene.

4. A process according to any one of the preceding claims, wherein the impurity is selected from metals, metal oxides and combinations thereof.

5. A process according to claim 4, wherein the impurity is selected from iron, elemental iron, iron oxides, iron-carbon species such as ferrite, austenite or cementite, and combinations thereof.

6. A process according to claim 4 or claim 5, wherein the iron impurity deposits as elemental iron on the cathode, precipitates as iron hydroxide or iron complex or remains in solution.

7. A process according to any one of the preceding claims, wherein the cathode comprises one or more metal, metal alloy plates, platinum, or titanium coated with platinum, and wherein the anode comprises graphite, lead, lead alloys, platinum, titanium coated with

platinum and combinations thereof.

8. A process according to claim 7, further comprising the use of a permeable membrane covering at least a portion of the anode, cathode, or both, wherein the permeable membrane is a neutrally-charged permeable membrane, an anion exchange membrane, or a cation exchange membrane, and wherein the permeable membrane has a molecular weight cut-off (MWCO) of less than about 1 million Da, and preferably between about 10 kDa, and about 0.5 kDa.

9. A process according to any one of the preceding claims, wherein the crude graphitic material is slurried prior to the electrochemical treatment process.

10. A purified graphitic material, obtained or obtainable from a process as defined according to any one of the preceding claims.

11. A negative electrode comprising a coating on a substrate, the coating comprising:

MnO_2 (electrolytic manganese dioxide (EMD)), a carbon conductive additive in the form of purified graphitic material according to claim 10, and a binder.

12. An electrode according to claim 11, wherein the EMD is one or more of α-, β-, γ-, δ-, or λ-MnO_2.

13. An electrode according to claim 11 or claim 12, wherein the EMD, the carbon conductive additive and the binder are mixed together in a weight ratio of 4-9:2:1.

14. A battery comprising:

a positive electrode;

a negative electrode according to any one of claims 11 to 13; and

an electrolyte in contact with the positive electrode and the negative electrode.

15. A battery according to claim 14, wherein the electrolyte is an aqueous electrolyte selected from the group consisting of an ammonium sulfate, sodium sulfate, magnesium sulfate, iron sulfate, copper sulfate, zinc sulfate ($ZnSO_4$), zinc chloride ($ZnCl_2$), zinc trifluoromethyl sulfonate ($Zn(CF_3SO_3)_2$), cadmium nitrate, cobalt nitrate, sodium nitrate, sodium chloride, nickel chloride, potassium chloride, ammonium chloride (NH_4Cl), calcium chloride, sulfuric acid, and combinations thereof; or wherein the electrolyte is an ionic liquid comprising a cation selected from the group consisting of 1-alkyl-3-methyl-

imidazolium, *N*-alkyl-pyridinium, tetraalkyl-ammonium, tetralkyl-phosphonium, 1-ethyl-3-methyl-1*H*-imidazolium, 1-butyl-3-methyl-1*H*-imidazolium, 1-butylpyridinium, and combinations thereof, wherein the alkyl group is selected from the group consisting of C_2-C_{12} alkyl .