15 V is applied". Lu et al. showed that the graphene nanosheet production is exclusively at the anode and is due to an interaction of decomposed water species and the anions from the ionic liquid, such as BF_4^-.

WO 2011/162727, discloses the formation of graphene using lithium ion exfoliation of graphite, the exfoliation being aided by the insertion of solvent between the layers and sonication. This work is also discussed in a related paper [Wang 2011].

The co-pending international application, filed on 9 March 2011 claiming priority from GB1104096.1 (filed 10 March 2011), entitled "Production of Graphene", describes the production of graphene by the electrochemical intercalation of alkylammonium ions into graphitic cathodes.

Further methods for the production of graphene are desired – in particular, methods that produce graphene sheets with a controlled number of layers and flake size. Advantageously, the methods should be scalable to allow for the production of graphene on a large scale.

Summary

In a first aspect, the present invention refers to a method for the production of graphite nanoplatelet structures having a thickness of less than 100 nm in an electrochemical cell, wherein the cell comprises:

(a) a negative electrode which is graphitic;

(b) a positive electrode which may be graphitic or another material; and

(c) an electrolyte which is ions in a solvent where the cations are organic ions and metal ions;

and wherein the method comprises the step of passing a current through the cell to produce the graphene and/or graphite nanoplatelet structures at the negative electrode.

Description of the Invention

The present inventors have conceived a method for the production of graphene and related graphite nanoplatelet structures by exfoliation driven by the electrochemical insertion of both positive organic ions (e.g. alkylammonium ions) and positive metal ions (e.g. iron, tin, lithium ions) into a negative graphitic electrode. Without wishing to be bound by theory, it is thought that by using cations to exfoliate the negative electrode, the possibility of the formation of graphene oxide through oxidative attack is reduced.

In the present application, the term "graphene" is used to describe materials consisting of ideally one to ten graphene layers, preferably where the distribution of the number of layers in the product is controlled. The method can also be used to make graphite nanoplatelet structures under 100 nm in thickness, preferably under 10 nm in thickness and more preferably under 1 nm in thickness. The size of the graphene flakes produced can vary from nanometres across to millimetres, depending on the morphology desired.

In some aspects of the invention, the material produced is graphene having up to ten layers. The graphene produced may have one, two, three, four, five, six, seven, eight, nine or ten layers. It may be preferred that the material produced is substantially free of graphene oxide.

Claims

1. A method for the production of graphene and graphite nanoplatelet structures having a thickness of less than 100 nm in an electrochemical cell, wherein the cell comprises:
(a) a negative electrode which is graphitic;
(b) a positive electrode which may be graphitic or another material; and
(c) an electrolyte which is ions in a solvent where the cations are organic ions and metal ions;
and wherein the method comprises the step of passing a current through the cell to produce the graphene and/or graphite nanoplatelet structures at the negative electrode.

2. A method according to claim 1, wherein the negative electrode comprises a layered graphite compound in which cations can be intercalated.

3. A method according to either claim 1 or claim 2, wherein the negative electrode is selected from highly ordered pyrolytic graphite, natural graphite and synthetic graphite.

4. A method according to any one of claims 1 to 3, wherein the organic cations are alkylammonium cations.

5. A method according to claim 4, where the alkylammonium cations are tetraalkyl ammonium.

6. A method according to claim 5, wherein the alkylammonium cations are selected from tetrabutyl ammonium, tetraethylammonium and tetramethyl ammonium.

7. A method according to claim 4, wherein the alkylammonium cations are trialkyl ammonium.

8. A method according to claim 7, wherein the alkylammonium cations are selected from tributyl ammonium, triethylammonium and trimethyl ammonium.

9. A method according to claim 4, wherein the alkylammonium cations are dialkyl ammonium.

10. A method according to claim 9, wherein the alkylammonium cations are selected from dibutyl ammonium, diethylammonium and dimethyl ammonium.