Claims

~~1. A composite material of a carbonaceous substance comprising a doped catalytic compound obtained by a sol-gel method comprising:~~

~~mixing a hydrolyzed solution comprising a precursor of a catalytic material with a carbonaceous material to obtain a sol;~~

~~incubating the sol under mixing;~~

~~condensating the sol to obtain a gel-coated composite;~~

~~subjecting the gel-coated composite to a first calcination carried out in an oxidizing environment; and~~

~~subjecting the calcined gel-coated composite to a second calcination carried out in a non-oxidizing environment, wherein the non-oxidizing environment comprises a second dopant comprising precursor material;~~

~~wherein a solution of a first dopant comprising precursor material is added to the solution comprising an organometallic precursor of a catalytic material before hydrolyzation, or the first dopant comprising precursor material is added to the gel-coated composite before subjecting the gel-coated composite to calcination.~~

~~2. The composite material of claim 1, further comprising a process of repeated coating, wherein the process comprises:~~

~~a) mixing a hydrolyzed solution comprising a precursor of a catalytic material and condensating it to obtain a gel;~~

~~b) adding a solution of a dopant comprising precursor material to the gel;~~

~~c) drying the gel-coated composite obtained directly after condensation but before calcination as referred to in claim 1;~~

~~d) mixing the gel obtained in b) with the dried gel-coated composite of c) to obtain a repeatedly coated composite material;~~

~~e) drying the repeatedly coated composite material;~~

~~f) subjecting the repeatedly coated composite material to a first calcination carried out in an oxidizing environment; and~~

~~g) subjecting the calcined repeatedly coated composite material to a second calcination carried out in a non-oxidizing environment.~~

~~3. The composite material of claim 2, wherein the process of repeated coating is carried out at least once using the dried repeatedly coated carbonaceous material obtained in a previous round under e) for mixing with the gel referred to under d) of claim 2.~~

~~4. The composite material of any one of the preceding claims, wherein the sol is heated for condensation.~~

~~5. The composite material of claim 4, wherein the sol is gradually heated for condensation.~~

~~6. The composite material of any one of the preceding claims, wherein the oxidizing environment is an oxygen comprising environment.~~

~~7. The composite material of any one of the preceding claims, wherein the non-oxidizing environment is an inert atmosphere.~~

~~8. The composite material of claim 7, wherein the inert atmosphere is nitrogen atmosphere or argon atmosphere.~~

~~9. The composite material of any one of the preceding claims, wherein the second dopant comprising precursor material is for the same or a different dopant than the first dopant comprising precursor material.~~

~~10. The composite material of any one of claims 7 to 9, wherein the second dopant comprising precursor material is comprised in the inert atmosphere in a mol% in relation to the inert gas of at most 50%.~~

~~11. The composite material of any one of the preceding claims, wherein the carbonaceous material is pre-treated with a basic solution.~~

~~12. The composite material of any one of the preceding claims, wherein the organometallic precursor is dissolved in an alcohol.~~

~~13. The composite material of any one of the preceding claims, wherein the first calcination is carried out at a temperature which is below the temperature used for the second calcination.~~

~~14. The composite material of any one of the preceding claims, wherein the first calcination is carried out at a temperature between about 400°C to about ≤500 °C.~~

~~15. The composite material of any one of the preceding claims, wherein the second calcination is carried out at a temperature between about > 500°C to about 700°C.~~

~~16. The composite material of any one of the preceding claims, wherein calcination is carried out for about 2 to 4 hours.~~

~~17. The composite material of any one of the preceding claims, wherein the first and second calcination are carried out independently of each other at a ramping rate of 5°C to 10°C per minute.~~

~~18. The composite material of any one of the preceding claims, wherein incubating the sol under mixing is carried out for a time period of between about 12 to 24 hours.~~

~~19. The composite material of any one of the preceding claims, wherein the carbonaceous material is a nanostructured carbonaceous material.~~

~~20. The composite material of any one of the preceding claims, wherein the carbonaceous material is activated carbon.~~

~~21. The composite material of any one of the preceding claims, wherein the activated carbon is in powder form.~~

~~22. The composite material of any one of the preceding claims, wherein the catalytic material is a photocatalytic material.~~

~~23. The composite material of claim 22, wherein the photocatalytic material is selected from the group consisting of TiO_2, ZnO, ZrO, CdS, MoS_2, Fe_2O_3, SnO_2, ZnS, W_2O_3, V_2O_5 and $SrTiO_3$.~~

~~24. The composite material of any one of the preceding claims, wherein the carbonaceous material is loaded with the doped catalytic material in an amount of between about 10 wt.% to about 50 wt.%.~~

~~25. The composite material of any one of the preceding claims, wherein the dopant is a non-metal dopant.~~

~~26. The composite material of claim 25, wherein the non-metal dopant is selected from the group consisting of nitrogen, halogen, sulfur, and phosphor.~~

~~27. The composite material of any one of the preceding claims, wherein the dopant comprising precursor material is selected from the group consisting of NH_3, urea, NH_4OH, hydrazine, amine, thiourea, carbon disulfide, iodic acid, sodium hypophosphite and hypophosphorous acid.~~

~~28. The composite material of any one of the preceding claims, wherein the precursor of a catalytic material is a metallic alkoxide or an organometallic precursor.~~

~~29. The composite material of any one of the preceding claims, wherein the molar ratio of precursor of a catalytic material to first dopant comprising precursor material is in the range of 1 : 10.~~

~~30. A method of removing pollutants comprised in a liquid stream by subjecting the liquid stream to a composite material of any one of claims 1 to 29.~~

~~31. The method of claim 30, wherein the liquid stream is flowing through a water treatment plant comprising a membrane filtration reactor.~~

~~32. The method of claim 31, wherein the pollutant is a recalcitrant pollutant.~~

~~33. The method of claim 32, wherein the recalcitrant pollutant is an organic pollutant.~~

~~34. The method of claim 33, wherein the organic pollutant is selected from the group consisting of a pharmaceutical composition, a plasticizer, a hormone, personal care product, disinfection by-product, surfactant; a perfluorinated chemical, a microcystin toxin, and natural organic matter (NOM).~~

~~35. A photocatalytic oxidation reactor comprising a composite material of any one of claims 1 to 29 wherein the composite material comprises a doped photocatalytic material.~~

~~36~~1. A sol-gel method for preparing a composite material of a carbonaceous substance comprising a doped catalytic compound, the method comprising:

mixing a hydrolyzed solution comprising a precursor of a catalytic material with a carbonaceous material to obtain a sol;

incubating the sol under mixing;

condensating the sol to obtain a gel-coated composite;

subjecting the gel-coated composite to a first calcination carried out in an oxidizing environment; and

subjecting the calcined gel-coated composite to a second calcination carried out in a non-oxidizing environment, wherein the non-oxidizing environment comprises a second dopant comprising precursor material;

wherein a solution of a first dopant comprising precursor material is added to the solution comprising an organometallic precursor of a catalytic material before hydrolyzation, or the first dopant comprising precursor material is added to the gel-coated composite before subjecting the gel-coated composite to calcination.

372. The method of claim 361, further comprising a process of repeated coating, wherein the process comprises:

a) mixing a hydrolyzed solution comprising a precursor of a catalytic material and condensating it to obtain a gel;

b) adding a solution of a dopant comprising precursor material to the gel;

c) drying the gel-coated composite obtained directly after condensation but before calcination as referred to in claim 36;

d) mixing the gel obtained in b) with the dried gel-coated composite of c) to obtain a repeatedly coated composite material;

e) drying the repeatedly coated composite material;

f) subjecting the repeatedly coated composite material to a first calcination carried out in an oxidizing environment; and

g) subjecting the calcined repeatedly coated composite material to a second calcination carried out in a non-oxidizing environment.

383. The method of claim 372, wherein the process of repeated coating is carried out at least once using the dried repeatedly coated carbonaceous material obtained in a previous round under e) for mixing with the gel referred to under d) of claim 37.

394. The method of any one of claims 36–1 to 383, wherein the sol is heated for condensation.

405. The method of claim 394, wherein the sol is gradually heated for condensation.

416. The method of any one of claims 36–1 to 405, wherein the oxidizing environment is an oxygen comprising environment.

427. The method of any one of the claims 36–1 to 416, wherein the non-oxidizing environment is an inert atmosphere.

438. The method of claim 427, wherein the inert atmosphere is nitrogen atmosphere or argon atmosphere.

449. The method of any one of the claims ~~36–~~1 to ~~43~~8, wherein the second dopant comprising precursor material is for the same or a different dopant than the first dopant comprising precursor material.

4510. The method of any one of claims ~~42–~~7 to ~~44~~9, wherein the second dopant comprising precursor material is comprised in the inert atmosphere in a mol% in relation to the inert gas of at most 50%.

4611. The method of any one of claims ~~36–~~1 to ~~45~~10, wherein the carbonaceous material is pre-treated with a basic solution.

4712. The method of any one of claims ~~36–~~1 to ~~46~~11, wherein the organometallic precursor is dissolved in an alcohol.

4813. The method of any one of claims ~~36–~~1 to ~~47~~12, wherein the first calcination is carried out at a temperature which is below the temperature used for the second calcination.

4914. The method of any one of claims ~~36–~~1 to ~~48~~13, wherein the first calcination is carried out at a temperature between about 400°C to about ≤500 °C.

5015. The method of any one of claims ~~36–~~1 to ~~49~~14, wherein the second calcination is carried out at a temperature between about > 500°C to about 700°C.

5116. The method of any one of claims ~~36–~~1 to ~~50~~15, wherein calcination is carried out for about 2 to 4 hours.

5217. The method of any one of claims ~~36–~~1 to ~~51~~16, wherein the first and second calcination are carried out independently of each other at a ramping rate of 5°C to 10°C per minute.

~~53~~18. The method of any one of claims ~~36~~-1 to ~~52~~17, wherein incubating the sol under mixing is carried out for a time period of between about 12 to 24 hours.

~~54~~19. The method of any one of claims ~~36~~-1 to ~~53~~18, wherein the carbonaceous material is a nanostructured carbonaceous material.

~~55~~20. The method of any one of claims ~~36~~-1 to ~~54~~19, wherein the carbonaceous material is activated carbon.

~~56~~21. The method of any one of claims ~~36~~-1 to ~~55~~20, wherein the activated carbon is in powder form.

~~57~~22. The method of any one of claims ~~36~~-1 to ~~56~~21, wherein the catalytic material is a photocatalytic material.

~~58~~23. The method of claim ~~57~~22, wherein the photocatalytic material is selected from the group consisting of TiO_2, ZnO, ZrO, CdS, MoS_2, Fe_2O_3, SnO_2, ZnS, W_2O_3, V_2O_5 and $SrTiO_3$.

~~59~~24. The method of any one of claims ~~36~~-1 to ~~58~~23, wherein the carbonaceous material is loaded with the doped catalytic material in an amount of between about 10 wt.% to about 50 wt.%.

~~60~~25. The method of any one of claims ~~36~~-1 to ~~59~~24, wherein the dopant is a non-metal dopant.

~~61~~26. The method of claim ~~60~~25, wherein the non-metal dopant is selected from the group consisting of nitrogen, halogen, sulfur, and phosphor.

~~62~~27. The method of any one of claims ~~36~~-1 to ~~61~~26, wherein the dopant comprising precursor material is selected from the group consisting of NH_3, urea, NH_4OH, hydrazine, amine, thiourea, carbon disulfide, iodic acid, sodium hypophosphite and hypophosphorous acid.

~~63~~28. The method of any one of claims ~~36~~ 1 to ~~62~~27, wherein the precursor of a catalytic material is a metallic alkoxide or an organometallic precursor.

~~64~~29. The method of any one of claims ~~36~~ 1 to ~~63~~28, wherein the molar ratio of precursor of a catalytic material to first dopant comprising precursor material is in the range of 1 : 10.

Claims

1. A sol-gel method for preparing a composite material of a carbonaceous substance comprising a doped catalytic compound, the method comprising:

mixing a hydrolyzed solution comprising a precursor of a catalytic material with a carbonaceous material to obtain a sol;

incubating the sol under mixing;

condensating the sol to obtain a gel-coated composite;

subjecting the gel-coated composite to a first calcination carried out in an oxidizing environment; and

subjecting the calcined gel-coated composite to a second calcination carried out in a non-oxidizing environment, wherein the non-oxidizing environment comprises a second dopant comprising precursor material;

wherein a solution of a first dopant comprising precursor material is added to the solution comprising an organometallic precursor of a catalytic material before hydrolyzation, or the first dopant comprising precursor material is added to the gel-coated composite before subjecting the gel-coated composite to calcination.

2. The method of claim 1, further comprising a process of repeated coating, wherein the process comprises:

a) mixing a hydrolyzed solution comprising a precursor of a catalytic material and condensating it to obtain a gel;

b) adding a solution of a dopant comprising precursor material to the gel;

c) drying the gel-coated composite obtained directly after condensation but before calcination as referred to in claim 36;

d) mixing the gel obtained in b) with the dried gel-coated composite of c) to obtain a repeatedly coated composite material;

e) drying the repeatedly coated composite material;

f) subjecting the repeatedly coated composite material to a first calcination carried out in an oxidizing environment; and

g) subjecting the calcined repeatedly coated composite material to a second calcination carried out in a non-oxidizing environment.

3. The method of claim 2, wherein the process of repeated coating is carried out at least once using the dried repeatedly coated carbonaceous material obtained in a previous round under e) for mixing with the gel referred to under d) of claim 37.

4. The method of any one of claims 1 to 3, wherein the sol is heated for condensation.

5. The method of claim 4, wherein the sol is gradually heated for condensation.

6. The method of any one of claims 1 to 5, wherein the oxidizing environment is an oxygen comprising environment.

7. The method of any one of the claims 1 to 6, wherein the non-oxidizing environment is an inert atmosphere.

8. The method of claim 7, wherein the inert atmosphere is nitrogen atmosphere or argon atmosphere.

9. The method of any one of the claims 1 to 8, wherein the second dopant comprising precursor material is for the same or a different dopant than the first dopant comprising precursor material.

10. The method of any one of claims 7 to 9, wherein the second dopant comprising precursor material is comprised in the inert atmosphere in a mol% in relation to the inert gas of at most 50%.

11. The method of any one of claims 1 to 10, wherein the carbonaceous material is pre-treated with a basic solution.

12. The method of any one of claims 1 to 11, wherein the organometallic precursor is dissolved in an alcohol.

13. The method of any one of claims 1 to 12, wherein the first calcination is carried out at a temperature which is below the temperature used for the second calcination.

14. The method of any one of claims 1 to 13, wherein the first calcination is carried out at a temperature between about 400°C to about ≤500 °C.

15. The method of any one of claims 1 to 14, wherein the second calcination is carried out at a temperature between about > 500°C to about 700°C.

16. The method of any one of claims 1 to 15, wherein calcination is carried out for about 2 to 4 hours.

17. The method of any one of claims 1 to 16, wherein the first and second calcination are carried out independently of each other at a ramping rate of 5°C to 10°C per minute.

18. The method of any one of claims 1 to 17, wherein incubating the sol under mixing is carried out for a time period of between about 12 to 24 hours.

19. The method of any one of claims 1 to 18, wherein the carbonaceous material is a nanostructured carbonaceous material.

20. The method of any one of claims 1 to 19, wherein the carbonaceous material is activated carbon.

21. The method of any one of claims 1 to 20, wherein the activated carbon is in powder form.

22. The method of any one of claims 1 to 21, wherein the catalytic material is a photocatalytic material.

23. The method of claim 22, wherein the photocatalytic material is selected from the group consisting of TiO_2, ZnO, ZrO, CdS, MoS_2, Fe_2O_3, SnO_2, ZnS, W_2O_3, V_2O_5 and $SrTiO_3$.

24. The method of any one of claims 1 to 23, wherein the carbonaceous material is loaded with the doped catalytic material in an amount of between about 10 wt.% to about 50 wt.%.

25. The method of any one of claims 1 to 24, wherein the dopant is a non-metal dopant.

26. The method of claim 25, wherein the non-metal dopant is selected from the group consisting of nitrogen, halogen, sulfur, and phosphor.

27. The method of any one of claims 1 to 26, wherein the dopant comprising precursor material is selected from the group consisting of NH_3, urea, NH_4OH, hydrazine, amine, thiourea, carbon disulfide, iodic acid, sodium hypophosphite and hypophosphorous acid.

28. The method of any one of claims 1 to 27, wherein the precursor of a catalytic material is a metallic alkoxide or an organometallic precursor.

29. The method of any one of claims 1 to 28, wherein the molar ratio of precursor of a catalytic material to first dopant comprising precursor material is in the range of 1 : 10.