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Method for attachment of silicon-containing compounds to a surface and for the synthesis of hypervalent silicon-compounds

5 The present invention relates to a method for inducing a hypervalent state within silicon-containing compounds by which they can be chemically attached to a surface or substrate and/or organized onto a surface of a substrate. The compounds when attached to or organised on the surface
10 may have different physical and/or chemical properties compared to the starting materials.

 The chemical attachment of silicon-containing compounds to surfaces is known to the skilled person. Conventionally,
15 this is achieved by contacting a suitable surface with a suitable silicon containing compound in the presence of an activator and heating the surface. The reaction between the surface and the silicon-containing compounds is relatively slow. An example of the attachment of siloxanes
20 to a polyester/cotton fabric using the application of heat is disclosed in US4417066.

 In a first aspect, the present invention provides a method for the chemical attachment of one or more silicon-
25 containing compounds to a substrate, the method comprising:

 providing one or more silicon-containing compounds selected from siloxane compounds, silanol compounds, silyl ether compounds, silanolate compounds, halosilane compounds, silatrane compounds, and silazane compounds;

30 contacting one or more of the silicon-containing compounds with a surface of a substrate having one or more nucleophilic sites thereon;

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and exposing the silicon-containing compounds and the surface to electromagnetic radiation having a frequency from 0.3 to 30 GHz.

5 In a second aspect, the present invention provides a method for creating an array of silicon-containing compounds on a surface of a substrate, the method comprising:

providing one or more silicon-containing compounds
10 selected from siloxane compounds, silanol compounds, silyl ether compounds, silanolate compounds, halosilane compounds, silatrane compounds, and silazane compounds;

contacting one or more of these silicon-containing compounds with a surface of a substrate;

15 and exposing the substrate and the silicon-containing compounds to electromagnetic radiation having a frequency of from 0.3 to 30 GHz. The substrate may have one or more nucleophilic sites on its surface or one or more compounds having nucleophilic groups, such as alcohols, may be present
20 on the surface.

In the first and second aspects, the surface of the substrate may be on the exterior or interior of the substrate. The substrate may, for example, be a porous
25 polymer matrix and the silicon-containing compounds may be arranged on or attached to the interior surfaces of the pores of the matrix.

In a third aspect, the present invention provides a
30 method for the synthesis of silicon-containing hypervalent compounds, the method comprising:

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providing one or more silicon-containing compounds selected from siloxane compounds, silanol compounds, silyl ether compounds, silanolate compounds, halosilane compounds, silatrane compounds, and silazane compounds;

5 optionally contacting the one or more silicon-containing compounds with a surface of a substrate;

and exposing the silicon-containing compounds and, if present, the substrate to electromagnetic radiation having a frequency of from 0.3 to 30 GHz. The substrate may have one
10 or more nucleophilic sites on its surface or one or more compounds having nucleophilic groups, such as alcohols, may be present, and if a substrate is present, they may be located on the surface of the surface.

15 In a fourth aspect, the present invention provides a substrate having one or more silicon-containing compounds on the surface thereof, wherein the one or more silicon-containing compounds have been attached to the surface by the method of the present invention.

20 In a fifth aspect, the present invention provides a substrate having one or more silicon-containing compounds on the surface thereof, wherein the one or more silicon-containing compounds have been organized into an array on
25 the surface by the method of the present invention.

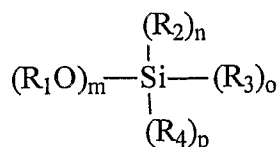
In a sixth aspect, the present invention provides a hypervalent silicon-containing compound formable by the method of the present invention.

30 The present invention will now be further described. In the following passages different aspects of then invention

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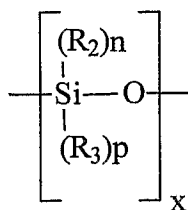
are defined in more detail. Each aspect so defined may be combined with any other aspect or aspects unless clearly indicated to the contrary. In particular, any feature indicated as being preferred or advantageous may be combined
 5 with any other feature or features indicated as being preferred or advantageous.

In the first, second and third aspects of the invention, the silicon-containing compound may be a compound
 10 of the formula I



formula I

or a polymer having repeating units of formula II, which may
 15 be terminated by hydrogen, hydroxyl, an alkyl or an amine group at one or both ends of the polymer chain



formula II

wherein R_1 is hydrogen or an alkyl, preferably a C_1 to
 20 C_6 alkyl, more preferably, a C_1 or a C_2 alkyl, such as methyl or ethyl, and m is 1 to 4, preferably 3;

R_2 , R_3 and R_4 are each independently selected from alkyl, alkylglycidoxo, alkylamino, aminoalkyl, acrylate, alkylhydantoin, alkylacrylate and alkylalkene; and

25 in formula I, n , m , o and p , are each 0 to 3, providing that $m+n+o+p = 4$;

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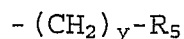
in formula II, n and p are each 0, 1 or 2, providing that $n + p = 2$.

The polymer preferably includes electron donor groups on at least some of its monomers. These electron donor groups may be substituents on R_2 and/or R_3 in the formula II above. Electron donor groups include, but are not limited to, hydroxyl, amine, sulfhydryl and carboxyl. X, the number of repeating units in the polymer, may be any appropriate number. X may be 2 to 10,000, preferably 2 to 1000.

Alkyl is preferably a C_1 to C_{25} alkyl, and may be C_3 to C_{18} alkyl. Alkyl may be a substituted or non-substituted alkyl. Alkyl may be a halo-alkyl, preferably a haloalkyl in which a halo group is located at the distal end of the alkyl chain from the silicon. The haloalkyl is preferably a chloroalkyl.

Preferably, the silicon-containing compound is a compound of formula I, wherein m is 3, n is 1 and o and p are both 0, R_1 is hydrogen, methyl or ethyl.

Preferably at least one of R_2 , R_3 and/or R_4 is of the formula III



formula III

wherein Y is 1 to 5, preferably 3, R_5 is selected from hydrogen, halogen, NH_2 , C_1 to C_{18} alkyl, C_1 to C_{18} alkyl dimethylammonium, alkylmethacrylate, preferably ethyl or propylmethacrylate, 5,5-dialkylhydantoin, preferably 5,5-dimethylhydantoin, alkylenediamine,

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preferably ethylenediamine, perfluoroalkyl, preferably perfluorooctyl and 3-glycidoxy.

The silicon-containing compound may comprise one or
5 more of [3-(trimethoxysilyl)propyl]-
octadecyldimethylammonium chloride, 3(3-
triethoxysilylpropyl)-5,5-dimethylhydantoin, potassium
trimethylsilanolate, triisopropylsilanol,
methoxydimethyloctylsilane, hydroxy terminated
10 poly(dimethylsiloxane), (3-chloropropyl)triethoxysilane,
(3-chloropropyl)dimethoxymethylsilane,
octadecyltrimethoxysilane, 3-aminopropyltriethoxysilane, 3-
(trimethoxysilyl)propylmethacrylate, N-[3-
(trimethoxysilyl)propyl]-ethylenediamine, and 3-
15 glycidoxypropyltrimethoxysilane, 1H,1H,2H,2H-
perfluorodecyltrimethoxysilane.

Preferably, the silicon-containing compound is [3-
(trimethoxysilyl)propyl]-octadecyldimethylammonium chloride
20 or 3(3-triethoxysilylpropyl)-5,5-dimethylhydantoin,
1H,1H,2H,2H-perfluorodecyltrimethoxysilane or a mixture
thereof.

In the first to third aspects of the invention, a
25 solution or suspension of the silicon containing compounds
may be contacted with the substrate. The solution and/or
suspension preferably comprises a polar solvent, more
preferably acetone and/or an alcohol, preferably both. The
alcohol preferably comprises methanol and/or ethanol.
30 Alternatively, the silicon-containing compound may be
solvent-free, i.e. not in the form of a solution or
suspension.

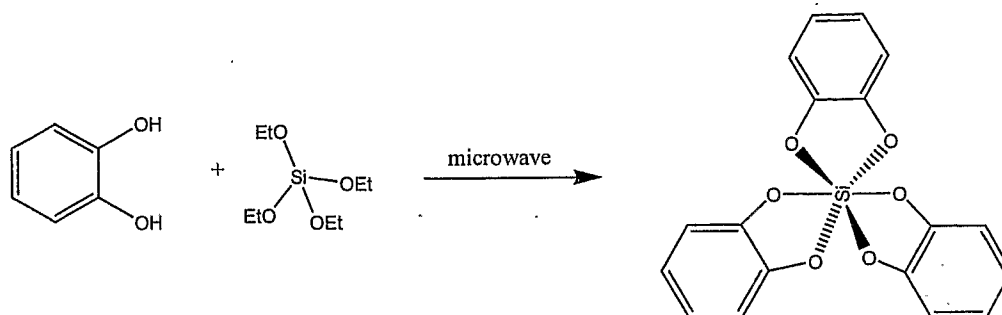
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The substrate is preferably a material having nucleophilic sites on its surface. The nucleophilic sites may comprise one or more nucleophilic groups containing one or more of O, S and N. For example, the nucleophilic groups may be selected from OH, SH and NH₂. The substrate may comprise a fabric material. It has been found that the nucleophilic groups bind to the silicon atoms of the silicon-containing compounds on contact and with exposure to electromagnetic radiation having a frequency of from 0.3 to 30 GHz. This reaction normally occurs within seconds, as opposed to hours for conventional methods, such as merely heating.

The present inventors have found that the method of the present invention can produce organised alignment (i.e. an array) of silicon-containing compounds on the surface of a substrate. The properties of the silicon-containing compounds often differ from those of the prior art, e.g. silicon compounds attached through the use of heat. For example, the silicon-containing compounds on the substrate may have different physical and chemical properties such as increased hydrophobicity or hydrophilicity and/or increased biocidal efficacy. Without being bound by theory, it is believed that the use of microwave radiation induces a hypervalency around the silicon atom, i.e. it coordinates to more than 4, possibly to 5 or 6 available ligands. When the microwave treatment is stopped, the silicon will often relax back to a tetra-coordinate state. The hypervalent state is believed to leads to a more organised arrangement of silicon-containing compounds on the surface of a substrate. Hypervalency in silicon has been shown to occur in the

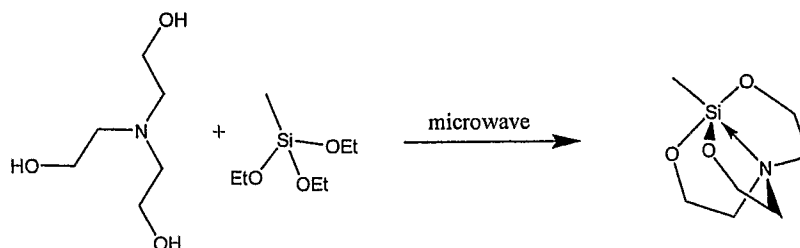
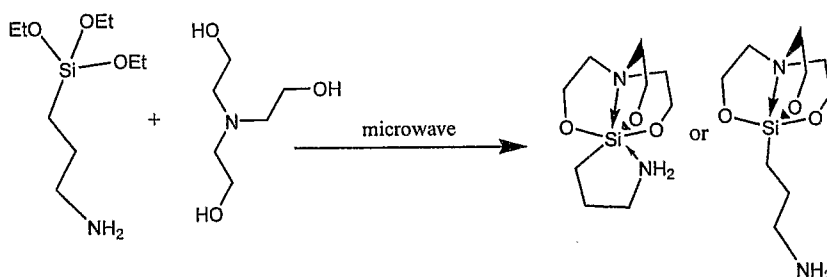
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following known reaction between tetramethoxy silane and catechol, which was carried out in ethanol under microwave treatment:



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Other reactions known to result in hypervalent-silicon compounds include:



15

The above reactions and others that result in hypervalent silicon can be found in the following prior art documents: *Chem. Rev.* **1993**, 93, 1371-1448, Chult et al;

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Chem. Rev. **1996**, *96*, 927-950, Holmes; and *Journal of Organometallic Chemistry*, **1990**, *389*, 159-168, Cerveau et al.

5 The hypervalent-silicon products produced in the above reactions are sufficiently stable to be characterised. It is believed that a similar hypervalency occurs in the process of the present invention, although the hypervalent silicon may revert to a tetravalent silicon following microwave treatment. It is surprising that, irrespective of
10 whether or not the silicon remains in a hypervalent state following the microwave treatment, a more organised arrangement of silicon-containing compounds on the surface of the substrate is observed.

15 Si-OR excitation via electromagnetic radiation in the presence of an appropriate electron donor facilitates the cleavage of the Si-OR bond and is believed to induce the formation of hypervalent siloxane species with available electron donors. The electron donor for this exchange can
20 take the form of virtually any nucleophile, induced nucleophile, nucleophilic region, or Lewis base. The resulting hypervalent species is then thought to either relax into their ground state, at which time the silane species is tetracoordinate, or, if the hypercoordinated
25 product is stable, the silane product can remain in the hypervalent state as either a pentacoordinate or hexacoordinate system. The electromagnetic excitation within the siloxane induces specific conformations within the new species, which leads to increased and specific
30 organization in the resting state of the newly formed species. If the substrate is a polymer, the specific organization of the silicon-containing on the surface or in

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the matrix of a polymer can change the chemical and physical properties of the polymer as a whole. This phenomenon is not temperature dependant.

5 The method of the present invention avoids the need to use activators, catalysts and conventional curing processes. This therefore allows the attachment of 'delicate' functionalities. For example, glycidoxo containing siloxane and acrylate containing siloxanes are examples of delicate
10 silicon containing compounds and proteins/enzymes are examples of delicate substrates, to which one may wish to attach a silicon-containing substrate.

 Preferably, the microwaves are produced using a power
15 rating of 650 Watts or less, more preferably of from 65 to 650 Watts. The microwaves may be produced using a power rating of from 135 to 400 Watts.

20 Preferably, the microwaves have a frequency of from 0.3 to 10 GHz, more preferably of from 1 to 3 GHz.

 To reduce the possible degradation of delicate silicon-containing compounds and/or delicate substrates, one or more
25 of the following may be used: irradiation at a reduced power level, for example microwaves produced using a power rating of 400 Watts or less, preferably 135 Watts or less, or subjecting the substrate and silicon-containing compounds to microwave irradiation and relaxation (no microwave
30 irradiation) in alternating intervals: for example a period of irradiation of preferably 5 to 30 seconds, more preferably 10 to 20 seconds, most preferably 15 seconds,

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followed by a period of relaxation of preferably 2 to 30 seconds, more preferably 5 to 15 seconds, most preferably 10 seconds, and optionally repeating this process as often as required. It has been found that, for many compounds
5 containing an Si-O moiety, this is more sensitive to microwave radiation than other 'delicate' functionalities and therefore cleavage of the Si-O bond may be achieved without degradation of the other functionalities. This is an improvement over the prior art in which heating of
10 silicon-containing compounds for periods to attach them to a substrate can lead to degradation of the delicate functionalities in the silicon-containing molecules, since the heat required to cleave the Si-O bond is sufficient to degrade the delicate functionalities.

15

The microwaves can be directed at particular portions of the substrate and therefore allows for regioselective attachment and/or arrangement of the silicon-substituted compounds and for reactions that can be initiated that would
20 not be possible using traditional methods.

25

The present invention has been found to be far more effective in attaching silicon-containing compounds to a substrate surface compared to traditional methods such as heating and using activators - more than 80% of the silicon-
containing compounds can be attached under certain conditions to the surface using the method of the present invention.

30

The substrate may comprise a natural material. The material may be a cloth material. The material may comprise one or more materials selected from cotton, wool and

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leather. The material may be woven or non-woven. The material may comprises fibres of natural and/or synthetic material. The synthetic material may comprise a woven or nonwoven fabric material to include, but not limited to
5 fabrics wherein the material comprises one or more of the following; cotton, polyester, nylon, wool, leather, rayon, polyethylene, polyvinylchloride, polyvinylalcohol, polyvinylamine and polyurea.

10 The substrate may be in the form of particles. The particles may have a diameter of from 10 nm to 1 mm, preferably 100 to 1000 nm.

The substrate may comprise a metal oxide. The metal
15 oxide may be selected from one or more of aluminium oxide, titanium dioxide, magnesium oxide, calcium oxide, silicon dioxide and zinc oxide.

The substrate may comprise a natural mineral. The
20 substrate may comprise one or more materials selected from kaolinite, barasym, silica, montmorillonite, vermiculite, bohemite and quartz.

The substrate may be porous. The substrate may
25 comprise a molecular sieve. The substrate may comprise a zeolite.

The substrate may comprise a polymer. The polymer may be in the form of a porous matrix. The substrate may
30 comprise a plastic material. The substrate may comprise polyurethane and/or nylon, polyester, nylon, rayon,

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polyethylene, polyvinylchloride, polyvinylalcohol, polyvinylamine and polyurea.

The substrate may comprise a carbohydrate.

5

In the first to third aspects of the present invention, an alcohol may be present. The substrate may comprise an alcohol. The substrate may have an alcohol on its surface. The alcohol may comprise a diol, which may be a vicinal
10 diol, or a triol. The alcohol may be selected from one or more of an alkyl diol, preferably a C₂ to C₂₅ alkyl diol, an alkyl triol, preferably a C₃ to C₂₅ alkyl triol and a phenyl diol, preferably a vicinal phenyl diol. Each hydroxyl group in the triol is preferably vicinal to one of the other
15 hydroxyl groups. The alcohol may be selected from catechol, ethylene glycol or glycerol.

The substrate may comprise a silicon-dioxide based material, such as glass, silicon dioxide, sand, and silica.

20

The method of the present invention may involve:

contacting the substrate with a silicon-containing compound as defined herein and exposing the compounds to electromagnetic radiation having a frequency of from 0.3 to
25 30 GHz, and subsequently treating the substrate with a halogenating substance. The silicon-containing compound preferably comprises 3(3-triethoxysilylpropyl)-5,5-dimethylhydantoin. The halogenating substance may comprise a chlorinating substance, including, but not limited to a
30 bleaching agent, for example a hypochlorite such as sodium hypochlorite.

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Following treating the substrate with the halogenating agent, the substrate may be dried. The substrate may be dried by exposing the substrate to a temperature of 20°C or more, preferably 30°C or more, more preferably 35°C for a
5 period including, but not limited to, 1 hour or more, preferably 4 hours or more.

The present invention will now be illustrated with reference to the following non-limited Examples.

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Examples**Example 1**

5 Separate swatches of cotton were each treated with a solution containing one of:

1. [3-(trimethoxysilyl)propyl]-octadecyldimethylammonium chloride,
- 10 2. 3(3-triethoxysilylpropyl)-5,5-dimethylhydantoin,
3. potassium trimethylsilanolate,
4. triisopropylsilanol,
5. methoxydimethyloctylsilane,
6. hydroxy terminated poly(dimethylsiloxane),
- 15 7. (3-chloropropyl)triethoxysilane,
8. (3-chloropropyl)dimethoxymethylsilane,
9. octadecyltrimethoxysilane,
10. 3-aminopropyltriethoxysilane,
11. 3-(trimethoxysilyl)propylmethacrylate,
- 20 12. N-[3-(trimethoxysilyl)propyl]-ethylenediamine, and
13. 3-glycidoxypropyltrimethoxysilane.

Each solution contained acetone, ethanol and methanol.

25 Each swatch of cotton was then irradiated at a frequency between 0.30-30 GHz for a period of time dependant on the properties of the silicon containing compound that was attached. All experiments were performed initially at 2.45 Ghz, with power level being varied depending on the

30 nature of the attached functional group. In general, the procedure was first attempted at full power for full two cycles of the following program: 30s 50% power (325 Watts),

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30s relaxation (magnetron disengaged), 30s 50% power, 30s relaxation, 30s 100% power, allow to cool to room temperature, then repeat cycle. If the procedure "cracked" the reactants, then the irradiation time and power level were reduced accordingly until the procedure yielded the desired result. The samples were then washed, rinsed thoroughly and dried overnight at 35° C.

The attachment of the silicon-containing compounds was confirmed by: 1. an overall increase in weight of the cloth (all samples), 2. a change in the physical characteristics of the cloth (all), 3. biocidal efficacy of the treated fabric (2 samples, see below), 4. FTIR spectroscopy (all), 5. elemental analysis (all), and 6. ionic strength of the product in distilled, deionized water (all).

Conclusions from Example 1: 1. the method is nonspecific with respect to the type of functionality that can be attached, and the compound may comprise a silyl ether, silanol, or silanolate; 2. The process works for silyl ethers, silanols and silanolates; and 3. by tuning the frequency and power level, the method can be tailored to avoid degradation of with other functionalities within the system; and 4. the process works on cotton.

Example 2

100 mg samples of nanoscale bohemite (300 nm from Sasol) were treated with one of the following solutions of:

1. [3-(trimethoxysilyl)propyl]-octadecyldimethylammonium chloride

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2. 3(3-triethoxysilylpropyl)-5,5-dimethylhydantoin,
3. potassium trimethylsilanolate,
4. triisopropylsilanol,
5. methoxydimethyloctylsilane,
- 5 6. hydroxy terminated poly(dimethylsiloxane),
7. (3-chloropropyl)triethoxysilane,
8. (3-chloropropyl)dimethoxymethylsilane,
9. octadecyltrimethoxysilane,
10. 3-aminopropyltriethoxysilane,
- 10 11. 3-(trimethoxysilyl)propylmethacrylate,
12. N-[3-(trimethoxysilyl)propyl]-ethylenediamine, and
13. 3-glycidoxypropyltrimethoxysilane.

Each solution contained one or more of acetone, ethanol
15 and methanol.

100 mg of bohemite is approximately 1 mmol. To this 1
mmol of bohemite approximately 0.1 mmol of silane dissolved
in approximately 0.5-1.0 mL of acetone was added. For
20 example, the MW of 3(3-triethoxysilylpropyl)-5,5-
dimethylhydantoin is 332, so 33.2 mg of silane was dissolved
in 1 mL. For the reactions above, the procedure was scaled
up to full molar scale. Surprisingly, the "batch" scale
actually worked much better than this small scale
25 experiment, i.e.. a higher and better degree of attachment
of the silicon on the bohemite was observed.

The samples were then irradiated at a frequency between
0.30-30 GHz for a period of time depending on the properties
30 of the silyl ether, silanolate or silanol that was attached.
The irradiation procedure described in Example 1 above was
used. The samples were then washed with a minimal amount of

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ice cold ethanol and then twice with surfactant-containing water; the samples were then rinsed with water thoroughly and dried overnight at 35° C.

5 The attachment of the silicon-containing compounds was confirmed by: 1. a change in the physical characteristics of the treated nanoparticles, 2. biocidal efficacy of the treated nanoparticles (2 samples, in particular: solutions 1 and 2), 4. FTIR spectroscopy (all), 5. elemental analysis
10 (all), and 6. ionic strength of the product in distilled, deionized water (all).

Conclusions from Example 2: 1. the process is nonspecific with respect to the type of functionality that
15 can be attached, and the silicon-containing compounds can include a silyl ether, silanol, or silanolate; 2. by tuning the frequency and power level, the process can be tailored not to interfere with other functionalities within the system; and 3. The process works well on bohemite

20

Example 3

Two samples of each of the substrates listed below were treated with either a solution of [3-
25 (trimethoxysilyl)propyl]-octadecyldimethylammonium chloride or a solution of 3(3-triethoxysilylpropyl)-5,5-dimethylhydantoin.

The substrate samples were: 1. Cotton, 2. aluminum
30 oxide, 3. titanium dioxide, 4. glass, 5. nylon, 6. kaolinite, 7. barasym, 8. silicon dioxide, 9. wool, 10. leather, 11. silica, 12. molecular sieves, 13.

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montmorillonite, 15. polyurethane, 16. ethylene glycol,
17. glycerol, 18. catechol, 19. zeolite, 20.
vermiculite, 21. bohemite, 22. polyester. 23.
triethanolamine

5

The samples were irradiated at a frequency between
0.30-30 GHz for a period of time, all dependant on the
microwave absorbing properties of the substrate. The
irradiation procedure described in Example 1 above was used.

10 The samples were then washed, rinsed thoroughly and dried
overnight at 35° C. The samples treated with 3(3-
triethoxysilylpropyl)-5,5-dimethylhydantoin) were also
charged with dilute hypochlorite (to generate the
chloramine) and again washed, rinsed and dried.

15

The attachment of the silicon-containing compounds to
each substrate was confirmed by:

1. a change in the physical characteristics of the
treated samples (all), 2. biocidal efficacy of the treated
20 samples (all solid samples), 3. oxidation of iodide to
elemental iodine (samples treated with 3(3-
triethoxysilylpropyl)-5,5-dimethylhydantoin), 4. FTIR
spectroscopy (all), 5. ionic strength of the product in
distilled, deionized water (all), 6. HNMR (2 samples: those
25 treated with Triethanolamine-hydantoin silane and catechol-
hydantoin silane)], 7. GCMS (3 samples: those treated with
catechol hydantoin silane, triethanolamine hydantoin silane
and glycerol hydantoin silane).

30 Conclusions from Example 3: 1. the process will work
for any substrate that contains an appropriate S, and/or O,
and/or N, and/or Nu; 2. by tuning the frequency and power

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level, the process can be tailored not to interfere with other functionalities within the system; and 3. the irradiated materials are distinctly different from the equivalent heat treated version (from HNMR and GCMS)

5

Example 4

Six swatches of 50:50 nylon:cotton cloth were each dipped one time in a known solution of 3(3-
10 triethoxysilylpropyl)-5,5-dimethylhydantoin. The first set of three swatch samples were irradiated at a frequency between 0.30-300 GHz for a period of time as described above in Example 1. This first set of samples was then chlorinated with dilute bleach, washed, rinsed thoroughly,
15 and dried overnight at 35° C. As a comparative Example, the second set of three swatch samples was heat cured overnight at 80° C. The second set of samples was then chlorinated with dilute bleach, washed, rinsed thoroughly, and dried overnight at 35° C. The attachment of the hydantoin
20 functionality of both sets of samples was confirmed by FTIR, and by iodometric titration. Results demonstrated that the irradiated samples retained 80% higher chlorine content over the heat treated samples. Additionally, the irradiated samples demonstrated hypervalent character by differences in
25 ionic strength.

Conclusions from Example 4: the irradiation process is more effective and efficient than heat treatment at attaching silyl ethers, silanols and silanolates to
30 substrates.

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Example 5

The biocidal character of heated treated samples of
5 50:50 cotton:nylon were compared with irradiated samples
from Example 4. Both sets of samples possessed the same
active chlorine content as determined by iodometric
titration. Results showed high excellent efficacy of
irradiated samples to *Bacillus anthracis* spores while heat
10 treated samples demonstrated minimal activity against
Bacillus anthracis spores.

Conclusions from Example 5: The irradiated materials
are distinctly different from the equivalent heat treated
15 version. The irradiation process produces a product that is
a more efficient sporicide than the heat cured process.

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Claims

1. A method for the chemical attachment of one or more silicon-containing compounds to a substrate, the method comprising:

providing one or more silicon-containing compounds selected from siloxane compounds, silanol compounds, silyl ether compounds, silanolate compounds, halosilane compounds, silatrane compounds, and silazane compounds;

contacting one or more of the silicon-containing compounds with a surface of a substrate having one or more nucleophilic sites thereon;

and exposing the silicon-containing compounds and the surface to electromagnetic radiation having a frequency from 0.3 to 30 GHz.

2. A method for creating an array of silicon-containing compounds on a surface of a substrate, the method comprising:

providing one or more silicon-containing compounds selected from siloxane compounds, silanol compounds, silyl ether compounds, silanolate compounds, halosilane compounds, silatrane compounds, and silazane compounds;

contacting one or more of these silicon-containing compounds with a surface of a substrate;

and exposing the substrate and the silicon-containing compounds to electromagnetic radiation having a frequency of from 0.3 to 30 GHz.

3. A method as claimed in claim 2, wherein the substrate has one or more nucleophilic sites on its surface or one or

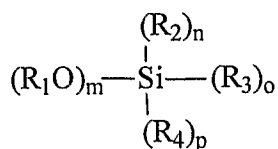
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more compounds having nucleophilic groups are present on the surface.

4. A method as claimed in claim 3, wherein the one or more
5 compounds having nucleophilic groups comprise an alcohol.

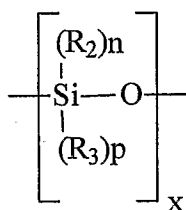
5. A method as claimed in any one of the preceding claims,
wherein the silicon-containing compound comprises a compound
of the formula I

10



formula I

or a polymer having repeating units of formula II,
which may be terminated by hydrogen or an alkyl group at one
15 or both ends of the polymer chain,



formula II

wherein R_1 is hydrogen or an alkyl, preferably a C_1 to
20 C_6 alkyl, more preferably, a C_1 or a C_2 alkyl, such as methyl
ethyl, and m is 1 to 3, preferably 3;

R_2 , R_3 and R_4 are each independently selected from
alkyl, alkylglycidoxo, alkylamino, aminoalkyl, acrylate,
alkylhydantoin, alkylacrylate and alkylalkene; and

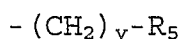
25 in formula I, n , m , o and p , are each 0 to 3, providing
that $m+n+o+p = 4$,

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in formula II, n and p are each 0, 1 or 2, providing that $n + p = 2$.

6. A method as claimed in claim 5, wherein the silicon-
5 containing compound is a compound of formula I, wherein m is 3, n is 1 and o and p are both 0, R_1 is hydrogen, methyl or ethyl.

7. A method as claimed in claim 5 or claim 6, wherein R_2 ,
10 R_3 and/or R_4 is of the formula III



formula III

wherein Y is 1 to 5, preferably 3, R_5 is selected from
15 hydrogen, halogen, NH_2 , C_1 to C_{18} alkyl,
 C_1 to C_{18} alkyldimethylammonium, alkylmethacrylate, preferably ethyl or propylmethacrylate, 5,5-dialkylhydantoin,
preferably 5,5-dimethylhydantoin, alkylenediamine,
preferably ethylenediamine, perfluoroalkyl, preferably
20 perfluorooctyl and 3-glycidoxy.

8. A method as claimed in any one of claims 1 to 4,
wherein the one or more silicon-containing compounds are
selected from [3-(trimethoxysilyl)propyl]-
25 octadecyldimethylammonium chloride, 3(3-
triethoxysilylpropyl)-5,5-dimethylhydantoin, potassium
trimethylsilanolate, triisopropylsilanol,
methoxydimethyloctylsilane, hydroxy terminated
poly(dimethylsiloxane), (3-chloropropyl)triethoxysilane,
30 (3-chloropropyl)dimethoxymethylsilane,
octadecyltrimethoxysilane, 3-aminopropyltriethoxysilane, 3-
(trimethoxysilyl)propylmethacrylate, N-[3-

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(trimethoxysilyl)propyl]-ethylenediamine, and 3-glycidoxypropyltrimethoxysilane, 1H,1H,2H,2H-perfluorodecyltrimethoxysilane.

- 5 9. A method as claimed in claim 8, wherein the one or more silicon-containing compound are selected from [3-(trimethoxysilyl)propyl]-octadecyldimethylammonium chloride and 3(3-triethoxysilylpropyl)-5,5-dimethylhydantoin.
- 10 10. A method as claimed in any one of the preceding claims, wherein the nucleophilic sites comprise one or more nucleophilic groups containing one or more of O, S and N.
11. A method as claimed in any one of the preceding claims,
15 wherein the substrate comprises a woven or nonwoven fabric material.
12. A method as claimed in claim 11, wherein the fabric material is selected from one or more of cotton, polyester,
20 nylon, wool, leather, rayon, polyethylene, polyvinylchloride, polyvinylalcohol, polyvinylamine and polyurea.
13. A method as claimed in any one of claims 1 to 10,
25 wherein the substrate comprises one or more materials selected from aluminum oxide, titanium dioxide, glass, nylon, kaolinite, barasym, silicon dioxide, silica, molecular sieves, montmorillonite, polyurethane, ethylene glycol, glycerol, catechol, zeolite, vermiculite, and
30 bohemite.

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14. A method as claimed in any one of the preceding claims, the method comprising contacting the substrate with 3(3-triethoxysilylpropyl)-5,5-dimethylhydantoin, exposing the compounds to electromagnetic radiation having a frequency of
5 from 0.3 to 30 GHz, and subsequently treating the substrate with a halogenating substance.

15. A substrate having a silicon-containing compound on a surface thereof, wherein the silicon-containing compound has
10 been attached to, and/or organised into an array on, the surface by the method as defined in any one of the preceding claims.

16. A method for the synthesis of hypervalent silicon-
15 containing compounds, the method comprising:

providing one or more silicon-containing compounds selected from siloxane compounds, silanol compounds, silyl ether compounds, silanolate compounds, halosilane compounds, silatrane compounds, and silazane compounds;

20 optionally contacting the one or more silicon-containing compounds with a surface of a substrate;

and exposing the silicon-containing compounds and, if present, the substrate to electromagnetic radiation having a frequency of from 0.3 to 30 GHz.

25

17. A hypervalent silicon-containing compound formable by the method as defined in any one of claims 1 to 14 and 16.