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(54) **Composite material for making sealing systems and method therefor**

(57) A method for making a composite material comprising a combination of biosoluble ceramics fibers and mineral fillers, for making sealing systems, comprising the steps of chemically pre-processing biosoluble ceramics fibers in an aqueous solution with vinyltriethoxysilane and mineral fillers with bis(triethoxysilyl-propyl) polysulfide or vice-versa and slowly mixing the processed biosoluble ceramics fibers and mineral fillers with an elastomeric solution as a process aid.

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Description

BACKGROUND OF THE INVENTION

[0001] The present invention relates to a composite material for making sealing systems and a method therefor.

[0002] In the thermal insulation field the use of ceramics fibers is well known owing to their optimum thermal and chemical resistance.

[0003] However, the use of conventional ceramics fibers is limited because of their suspected toxicity.

[0004] For the above reasons, biosoluble ceramics fibers being characterized as untoxic system have been developed.

[0005] In the present invention biosoluble ceramics fibers which will be hereinbelow referred to as "A" composite are exclusively used.

[0006] It is also known that mineral fillers have a sheet like microstructure and tend to overlap thereby providing structures which could be called "paling or palisade" constructions.

[0007] The above composite materials, which will be hereinbelow referred to as "B composites", comprise: muscovite and phlogophite mica, bentonite, montmorilonite, laponite, hydrotalcite, kaolin, lamellar structure silicates, phyllosilicates, thermally, ultrasonically and/or chemically foamed phlogophite mica.

[0008] Said B composites may also be used in any combinations and rates.

[0009] The A composite may be used as a fibrous reinforcement agent for composite materials, owing to its good mechanical and chemical properties; however, in the sealing or gasket system field it cannot be used per se, because of its poor sealing capability.

[0010] In turn, the B composites may be used in the sealing system field, since they provide good sealing capabilities, but cannot be used per se, because of their poor mechanical characteristics.

[0011] Moreover, a simple composite A and B mixture cannot be used since the individual composites thereof are incompatible with one another and do not form blended systems of continuous and homogeneous properties.

SUMMARY OF THE INVENTION

[0012] Thus, the aim of the present invention is to provide a combined composite material the A composite of which is chemically bound to the B composite, through an organosilane based chemical process.

[0013] Moreover, the end product appears as consisting of an uneven weave of biosoluble ceramics fibers operating as structural support for inorganic compounds having a layered lamellar structure.

[0014] The invention also relates to a method for making plates, films and sheets even coupled to other materials such as metal and non-metal fiber fabrics, smooth, perforated and/or diamond shaped metal laminates, which will be hereinbelow referred to as "supports".

[0015] The above aim and objects, as well as yet other objects which will become more apparent hereinafter, are achieved by a method for making a composite material, as well as the related composite material consisting of biosoluble ceramics fibers and mineral fillers, particularly for making sealing or tightness systems.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

[0016] Further characteristics and advantages of the present invention will become more apparent hereinafter from the following disclosure of a preferred, though not exclusive, embodiment of the invention.

[0017] The method according to the present invention comprises the following method steps:

A step 1) of chemically pre-processing biosoluble ceramics fibers (A composite) in an aqueous solution with vinyltriethoxysilane and mineral fillers (B composite) with bis(triethoxysilylpropyl)polysulfide or vice-versa.

[0018] The B composite may comprise muscovite and phlogophite mica, bentonite, montmorilonite, laponite, hydrotalcite, kaolin, lamellar structure silicates, phyllosilicates, thermally, ultrasonically and/or chemically foamed phlogophite mica.

[0019] A step 2) of preferably slowly mixing the already processed A and B composites, with an elastomeric material solution as a process aid.

[0020] A step 3) of forming a plate and/or a coupling with the supports.

[0021] A step 4) of finally thermally curing the material from step 3).

[0022] A step 5) of preferably hot coating the material surface by an anti-adhering protective film having a thickness larger than 5 μm, based on high temperature silica and PET, by a vacuum heated calender.

[0023] In the step 1) the A composite is processed with vinyltriethoxysilane in an aqueous or organic solution and with a concentration from 0.1 to 30% by weight, in which vinyltriethoxysilane is present starting from a rate from 0.5% up to

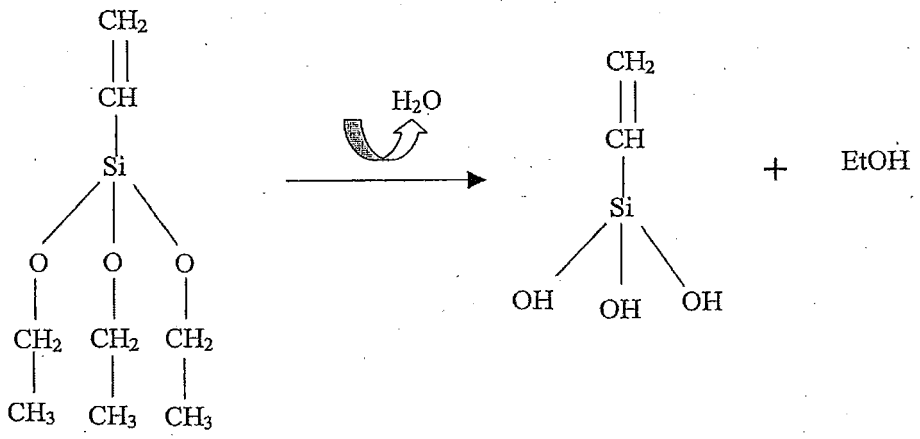
30% by weight, based on the A composite.

[0024] The B composite is processed with bis(triethoxy-silylpropyl)polysulfide in an aqueous or organic solution at a concentration from 0.1 to 60% by weight, wherein said bis(triethoxysilylpropyl)polysulfide is present from a rate of 0.5% to 30% by weight based on the B composite.

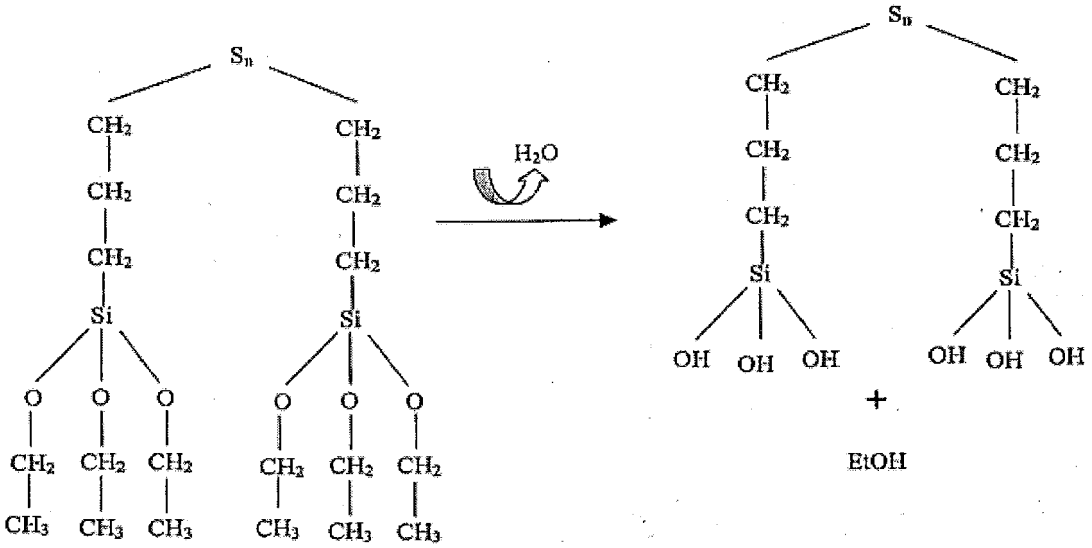
[0025] The above processes are carried out separately for the A and B composites, in a closed vessel and under slow stirring, at a temperature from 25°C to 80°C for 24 hours.

[0026] In this first step, the following chemical reaction occurs:

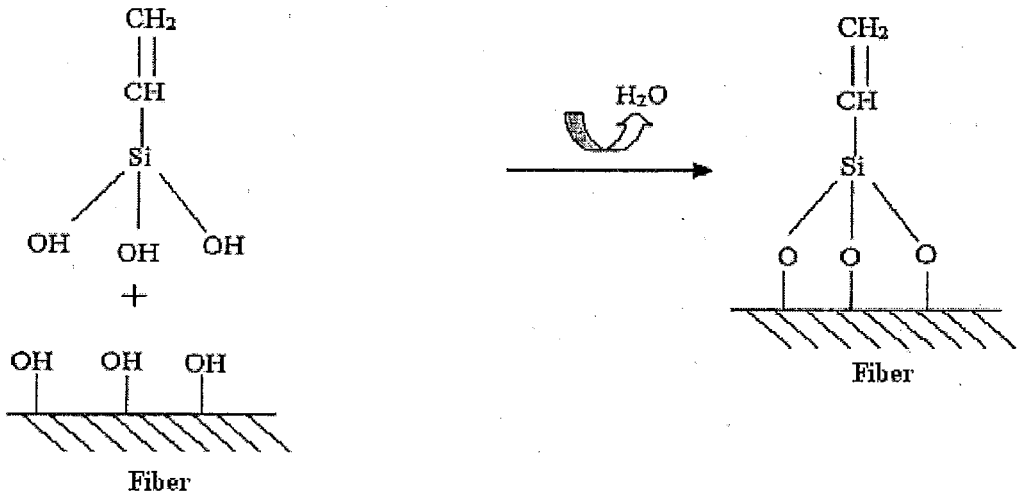
a) a vinyltrietoxysilane hydrolysis and an ethanol evolution



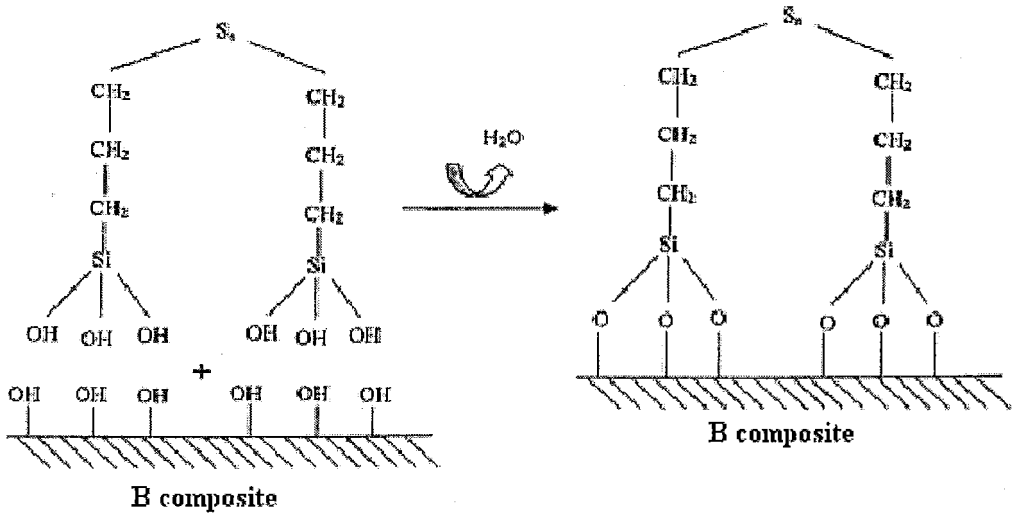
a bis(triethoxysilylpropyl)polysulfide hydrolysis and an ethanol evolution



b) a condensation reaction of the A composite surface hydroxyls and the vinyltrihydroxysilane hydroxyls



[0027] A condensation reaction between the B composite surface hydroxyls and the bis(trihydroxysilylpropyl)polysulfide hydroxyls



[0028] In said step 2) the A and B composites, as preprocessed in said step 1), are mixed with a solution polymer based binding system such as: NBR, chloroprene, SBR, polyisoprene, polybutadiene, butyl, silicone, fluorurated and urethane rubbers.

[0029] Preferably, the following solvents are herein used: toluene, xylene, dimethylketone methylethylketone, saturated C6-C10 linear and cyclic hydrocarbons.

[0030] The solutions contain the polymer in a rate up to 30% by weight.

[0031] The preferred mixing system comprises a "Z branch" or a planetary system.

[0032] The mixing time is preferably from 30 to 45 minutes, depending on the desired end viscosity.

[0033] The A composite may be introduced into the mixture in a rate up to 80% by weight.

[0034] The B composite may be introduced into the mixture in a rate up to 90% by weight.

[0035] This difference will depend on the required technical performance.

[0036] The process aid is introduced into the mixture in a rate up to 7% by weight.

[0037] As the A composite amount increases, a viscous creeping reduction in the gasket made from the plate is detected.

[0038] As the B composite amount increases, a decrease of the fluid loss (leakage) rate through the gasket made from the plate is detected.

[0039] In said step 3) the mixture of step 2) may be processed through calendaring or spreading and optionally coupled to a support, if either plates or continuous films are to be achieved.

[0040] In said step 4), the half-finished articles of step 3) are subjected to a thermal cycle up to 250°C depending on the used process aid polymer and solvent, with a heating rate from 1°C/minute.

[0041] The main reaction herein occurring are:

a) a breaking of the polysulfide residue;
b) a cross-linking reaction.

Examples:

Formulation 1

[0042]

1) Treating 10 kg of biosoluble fiber with 100 g vinyltriethoxysilane in ethanol at 5%, in a mixer for 12-24 hours, while processing 5 kg muscovite mica, with a particle size from 10 to 50 μm and 1 kg foamed phlogophite mica with a particle size from 200 to 500 μm, with 150 g bis(triethoxysilylpropyl)polysulfide in ethanol at 5% in a mixer for 12-24 hours.
2) Drying the processed A and B composites, in a dryer, with a solvent abatement system for 4 hours at 50°C, in steel basins or vats.
3) Mixing the dried A and B composites with 200 g NBR in a toluene solution at 20% and 1.8 kg deionized water, in a planetary mixer for a minimum time of 20 minutes.
4) Depositing the mixture of step 3) on a stainless steel 321 grid metal support, by a calendaring process.
5) Thermally processing in an oven at 180°C for 180 minutes.
6) Coating the sheet element by a PET and silica protective film with a thickness larger than 5 μm.

[0043] It has been found that the invention fully achieves the intended aim and objects.
[0044] In fact, the invention has provided a method of making a composite material, particularly suitable for making sealing systems, comprising a combination of biosoluble ceramics fibers and mineral filers.
[0045] The method according to the present invention provides an end product consisting of an uneven weave of biosoluble ceramics fibers, operating as a structural support for inorganic composites having a lamellar layered structure.
[0046] The sealing system composite material according to the present invention comprises a biosoluble fiber matrix having a thermal resistance up to 1000°C, with lamellar layered structure inorganic composites having a particle size from 1 μm.
[0047] The use of organosilanes in the subject method provides a mechanically continuous phase between the biosoluble fiber structure and the lamellar structure inorganic composites.
[0048] Moreover, the method according to the present invention provides semifinished sheets, plates, films or sheet elements coupled to other materials such as metal and non-metal fiber fabrics, smooth, perforated and/or diamond shaped metal laminates and other articles.
[0049] The composite material made by the present invention may be used at high temperatures up to 1,000°C, even in the presence of strongly oxidizing agents.
[0050] Moreover, the inventive composite material may also be used for high temperature sealing systems.
[0051] Furthermore, the inventive composite material may also be used as a protective film coated on surfaces of the sheet elements.

Claims

1. A method for making a composite material comprising a combination of biosoluble ceramics fibers and mineral fillers, for making sealing systems, **characterized in that** said method comprises the steps of:

   chemically pre-processing biosoluble ceramics fibers in an aqueous solution with vinyltriethoxysilane and mineral fillers with bis(triethoxysilylpropyl)-polysulfide or vice versa;
   slowly mixing the processed biosoluble ceramics fibers and mineral fillers with an elastomeric solution as a process aid.

2. A method, according to claim 1, **characterized in that** said mineral fillers comprise one or more of the following elements: mica, muscovite and phlogophite, bentonite, montmorilonite, laponite, hydrotalcite, kaolin, lamellar structure silicates, phyllosilicates, thermally, ultrasonically and/or chemically foamed phlogophite mica.

3. A method, according to claim 1, **characterized in that** said method comprises a further step of forming a plate and/or a laminate structure with the supports.

4. A method, according to claim 1, **characterized in that** said method comprises a further thermally curing end step.

5. A method, according to claim 1, **characterized in that** said method comprises a further step of hot coating a surface with an anti-adhering protective film having a thickness larger than 5μm, based on high temperature silica and PET, by a vacuum heated calendaring device.

6. A method, according to claim 1, **characterized in that** said method comprises processing the biosoluble ceramics fibers by vinyltriethoxysilane in an aqueous or organic solution at a concentration from 0.1 to 30% by weight, said vinyltriethoxysilane being present starting from an amount from 0.5% to 30% by weight based on the biosoluble ceramics fibers, and processing the mineral fillers by bis(triethoxysilyl-propyl)polysulfide in an aqueous or organic solution at a concentration from 0.1 to 60% by weight, said bis(triethoxysilylpropyl)polysulfide being present in an amount from 0.5% to 30% by weight with respect to said mineral fillers, said processing being carried out separately in a closed processing vessel and under a slow stirring at a temperature from 25°C to 80°C for 24 hours.

7. A method, according to claim 1, **characterized in that** said binding system is used with a process aid based on a solution polymer such as: NBR, chloroprene, SBR, polyisoprene, polybutadiene, butyl, silicone, fluorurated and urethane rubbers.

8. A method, according to claim 1, **characterized in that** in said method solvents selected from toluene, xylene, dimethylketone, methylethylketone, saturated C6-C10 linear and cyclic hydrocarbons are used.

9. A composite material for making sealing systems, **characterized in that** said composite material comprises a matrix consisting of an uneven weave of biosoluble fibers, operating as a support for lamellar layered structure inorganic composites.

10. A composite material, according to claim 9, **characterized in that** said biosoluble fiber matrix has a thermal resistance up to 1,000°C and said lamellar layered structure inorganic composites have a particle size from 1 μm.

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EUROPEAN SEARCH REPORT

Application Number
EP 12 19 1325

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (IPC)
A	US 5 814 271 A (BUJALSKI DUANE RAY [US] ET AL) 29 September 1998 (1998-09-29) * column 1, line 50 - column 3, line 46 * -----	1-10	INV. D04H1/58 C04B35/56 C04B35/622
A	EP 1 323 687 A2 (KUMKANG KOREA CHEM CO LTD [KR]) 2 July 2003 (2003-07-02) * paragraphs [0020], [0022]; claims 1,4 * -----	1-10	D04H1/4209
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TECHNICAL FIELDS SEARCHED (IPC)

D04H
C04B

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
Munich	30 April 2013	Lanniel, Geneviève

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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ANNEX TO THE EUROPEAN SEARCH REPORT ON EUROPEAN PATENT APPLICATION NO. EP 12 19 1325

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