(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
12 December 2024 (12.12.2024)

WIPO | PCT

(10) International Publication Number
WO 2024/253783 A1

(51) International Patent Classification:
D04H 1/413 (2012.01) *D04H 1/64* (2012.01)
B01J 13/00 (2006.01) *D06M 11/79* (2006.01)
C01B 33/154 (2006.01) *E04B 1/76* (2006.01)
C01B 33/158 (2006.01) *E04B 1/78* (2006.01)
C04B 14/06 (2006.01) *F16L 59/02* (2006.01)
D04H 1/587 (2012.01)

(21) International Application Number:
PCT/US2024/027929

(22) International Filing Date:
06 May 2024 (06.05.2024)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
202310671022.X 07 June 2023 (07.06.2023) CN

(71) Applicant: **OWENS CORNING INTELLECTUAL CAPITAL, LLC** [US/US]; One Owens Corning Parkway, Toledo, Ohio 43659 (US).

(72) Inventors: **LI, Hongze**; No. 118, Hongda Road, Qianjiang Economic Development Zone Linping Area, Hangzhou, Zhejiang (CN). **LI, Zheng**; No. 118, Hongda Road, Qianjiang Economic Development Zone Linping Area, Hangzhou, Zhejiang (CN). **QUE, Wenbin**; No. 118, Hongda Road, Qianjiang Economic Development Zone Linping Area, Hangzhou, Zhejiang (CN).

(74) Agent: **RAULERSON, Billy C.**; Calfee, Halter & Griswold LLP, The Calfee Building, 1405 East Sixth Street, Cleveland, Ohio 44114 (US).

(81) Designated States *(unless otherwise indicated, for every kind of national protection available)*: AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CV, CZ, DE, DJ, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IQ, IR, IS, IT, JM, JO, JP, KE, KG, KH, KN, KP, KR, KW, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, MG, MK, MN, MU, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, WS, ZA, ZM, ZW.

(84) Designated States *(unless otherwise indicated, for every kind of regional protection available)*: ARIPO (BW, CV, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SC, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT,

(54) Title: SILICON DIOXIDE AEROGEL COMPOSITE NON-WOVEN HEAT INSULATION SHEET

(57) Abstract: The present application provides a silicon dioxide aerogel composite non-woven heat insulation sheet, comprising a non-woven and silicon dioxide aerogel. The non-woven comprises a fiber and a binder bonding the fiber together. The binder has a hydroxyl group. The silicon dioxide aerogel is compounded on the non-woven. The silicon dioxide aerogel composite non-woven heat insulation sheet is obtained through a sol preparation process, a gel substrate composite process, a gel reaction process and a drying process, wherein the sol preparation process includes heating reflux at a first preset time at a first preset temperature after a catalyst, solvent and silicon source are mixed and stirred evenly and adding and mixing evenly a gel accelerator to form a sol precursor after cooling. The gel substrate composite process includes compounding the sol precursor into the non-woven. The gel reaction process includes causing gel aging of a non-woven compounded with the sol precursor at a second preset temperature for a second preset time.

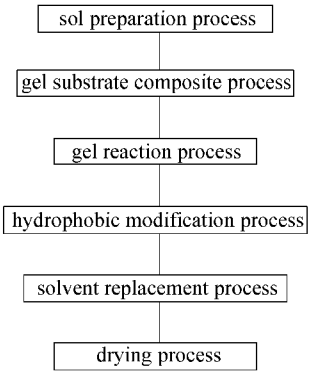

FIG. 1

WO 2024/253783 A1

[Continued on next page]

LU, LV, MC, ME, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

Published:

— *with international search report (Art. 21(3))*

SILICON DIOXIDE AEROGEL COMPOSITE NON-WOVEN HEAT INSULATION SHEET

CROSS-REFERENCE TO RELATED APPLICATION(S)

[0001] This application claims priority to Chinese Patent Application No. 202310671022.X filed on June 7, 2023, the disclosure of which is incorporated by reference herein in its entirety.

BACKGROUND

[0002] The present application relates to the technical field of composite materials, and specifically to a silicon dioxide aerogel composite non-woven heat insulation sheet.

[0003] Aerogel is a material of an extremely low density, which can be as low as 3kg/m^3, and also has a unique three-dimensional nanonetwork structure, large specific surface area and high porosity. Therefore, aerogel has excellent thermal insulation property and thereby widespread application prospects, as currently recognized solid material with the lowest thermal conductivity.

[0004] With the use of thermal insulation properties of aerogel, for example, the aerogel is combined with a fiber felt to form a thermal insulation felt, which has wide applications in fields such as aviation.

[0005] However, there are generally many problems in the preparation of the current thermal insulation felt. For example, as an inorganic chemical, the silicon dioxide aerogel is difficult to be bonded to the substrate by organic adhesives. Therefore, the powder loss problem of the silicon dioxide aerogel composite heat insulation sheet is always a pain spot in the industry.

[0006] Therefore, a silicon dioxide aerogel composite non-woven heat insulation sheet is needed to at least partially solve the above problems.

SUMMARY

[0007] A series of illustrative concepts is introduced into the portion of Summary, which would be further illustrated in the portion of the detailed description. The Summary of the present application does not mean attempting to define the key feature and essential technical feature of the claimed technical solution, let alone determining the protection scope thereof.

[0008] To at least partially solve the problems, the present application provides a silicon dioxide aerogel composite non-woven heat insulation sheet, comprising:

[0009] a non-woven comprising a fiber and a binder bonding the fiber together, the binder having a hydroxyl group;

[0010] silicon dioxide aerogel compounded on the non-woven;

[0011] the silicon dioxide aerogel composite non-woven heat insulation sheet obtained through a sol preparation process, a gel substrate composite process, a gel reaction process and a

drying process,

[0012] wherein the sol preparation process includes heating reflux at a first preset time at a first preset temperature after a catalyst, solvent and silicon source are mixed and stirred evenly and adding and mixing evenly a gel accelerator to form a sol precursor after cooling;

[0013] the gel substrate composite process includes compounding the sol precursor into the non-woven;

[0014] the gel reaction process includes causing gel aging of a non-woven compounded with the sol precursor at a second preset temperature for a second preset time.

[0015] According to the silicon dioxide aerogel composite non-woven heat insulation sheet of the present application, the binder of the non-woven contains the hydroxyl group, which can reduce the powder loss rate of products.

[0016] Optionally, the binder contains polyvinyl alcohol; and/or

[0017] the binder includes a reactant of polyacrylic acid and polyol. According to the above settings, the binder of the non-woven contains polyvinyl alcohol and/or the reactant of polyacrylic acid and polyol, which can effectively reduce the powder loss rate of products.

[0018] Optionally, the binder is polyvinyl alcohol.

[0019] According to the above settings, the use of PVA by the binder of the non-woven can greatly reduce the powder loss rate of products under a wide range of conditions.

[0020] Optionally, the catalyst is hydrochloric acid. The solvent comprises water and anhydrous ethanol. The silicon source is tetraethyl orthosilicate. The gel accelerator is ammonia water.

[0021] Optionally, the sol preparation process includes adding hydrochloric acid and anhydrous ethanol after being mixed into tetraethyl orthosilicate and stirring for 2-5min, heating reflux mixed solution at 80-90°C for 3-5h, cooling to 15-25°C, and then adding ammonia water diluted with anhydrous ethanol and stirring for 15-25min to obtain the sol precursor.

[0022] Optionally, the gel reaction process includes causing gel aging of a nonwoven compounded with the sol precursor for 20-28h at 30-40°C.

[0023] Optionally, a solvent replacement process is further included between the gel reaction process and the drying process, comprising:

[0024] treating a gel aged sheet with anhydrous ethanol at 50-60°C for 8-24h to replace all solvents with anhydrous ethanol.

[0025] Optionally, a hydrophobic modification process is further included between the gel reaction process and the solvent replacement process, comprising:

[0026] modifying a gel aged sheet with ethanol solution of silane of 20-30% volume fraction at 50-60°C for 8-24h.

[0027] Optionally, the drying process is CO_2 supercritical drying.

[0028] Optionally, the drying process is performed using a CO_2 supercritical drying device with a drying kettle and a separating kettle, comprising:

[0029] a gel aged sheet is placed in the drying kettle and CO_2 is injected into the CO_2 supercritical drying device with a flow rate of 35-40 L/h for 6-10h;

[0030] wherein the temperature in the drying kettle is controlled to be 50-70°C, and the pressure therein is controlled to be 17-18 MPa, so as to make CO_2 enter a supercritical state and replace ethanol in the gel aged sheet;

[0031] the temperature in the separating kettle is controlled to be 30-50°C and the pressure therein is controlled to be 7-8 MPa such that CO_2 and ethanol entering the separating kettle can be separated.

[0032] Optionally, the gel substrate composite process includes pouring the sol precursor into the non-woven by a perfusion process.

[0033] Optionally, the binder in the non-woven has a mass ratio of 5-30%.

[0034] Optionally, in the silicon dioxide aerogel composite non-woven heat insulation sheet, a ratio of the mass of the silicon dioxide aerogel to the mass of the non-woven is 5%-60%.

[0035] Optionally, the silicon dioxide aerogel composite non-woven heat insulation sheet has an air permeability of 1000-6000 mm/s.

[0036] Optionally, the silicon dioxide aerogel composite non-woven heat insulation sheet has a power loss rate of less than 1.5%.

[0037] Optionally, the silicon dioxide aerogel composite non-woven heat insulation sheet has a heat conductivity coefficient of less than 0.021 W/m•K.

[0038] Optionally, the silicon dioxide aerogel composite non-woven heat insulation sheet has a thickness of 0.3-3mm.

[0039] Optionally, the silicon dioxide aerogel composite non-woven heat insulation sheet has an areal density of 30-300g/m^2.

[0040] The second aspect of the present application provides a silicon dioxide aerogel composite non-woven heat insulation sheet, comprising:

[0041] a non-woven comprising a fiber and a binder bonding the fiber together, the binder having a hydroxyl group;

[0042] silicon dioxide aerogel compounded on the non-woven.

[0043] According to the silicon dioxide aerogel composite non-woven heat insulation sheet of the present application, the binder of the non-woven contains the hydroxyl group, which can reduce the powder loss rate of products.

[0044] Optionally, the binder contains polyvinyl alcohol; and/or

[0045] the binder includes a reactant of polyacrylic acid and polyol.

[0046] Optionally, the binder is polyvinyl alcohol.

[0047] Optionally, the binder in the non-woven has a mass ratio of 5-30%.

[0048] Optionally, in the silicon dioxide aerogel composite non-woven heat insulation sheet, a ratio of the mass of the silicon dioxide aerogel to the mass of the non-woven is 5%-60%.

[0049] Optionally, the silicon dioxide aerogel composite non-woven heat insulation sheet has an air permeability of 1000-6000 mm/s.

[0050] Optionally, the silicon dioxide aerogel composite non-woven heat insulation sheet has a power loss rate of less than 1.5%.

[0051] Optionally, the silicon dioxide aerogel composite non-woven heat insulation sheet has a heat conductivity coefficient of less than 0.021 W/m•K.

[0052] Optionally, the silicon dioxide aerogel composite non-woven heat insulation sheet has a thickness of 0.3-3mm.

[0053] Optionally, the silicon dioxide aerogel composite non-woven heat insulation has an areal density of 30-300g/m^2.

BRIEF DESCRIPTION OF THE DRAWINGS

[0054] The following drawings are hereby incorporated as part of the present application for the understanding of the application. The embodiments are illustrated and described in the drawings in order to explain the principles of the present application.

[0055] In the drawings:

[0056] FIG. 1 shows a schematic view of the preparation process of a silicon dioxide aerogel composite non-woven heat insulation sheet according to an embodiment of the present application; and

[0057] FIG. 2 shows a schematic view of a supercritical drying device according to an embodiment of the present application.

DETAILED DESCRIPTION

[0058] In the following description, numerous specific details are set forth in order to provide a more thorough understanding of the present application. However, it is obvious to those skilled in this art that the present application may be implemented without one or more of these details. Some technical features well-known in this art are not described in other examples in order to avoid confusion with the present application.

[0059] It shall be noted that the terminology used herein is for the purpose of describing particular embodiments only and is not intended to limit the exemplary embodiments of the present application. As used herein, the singular forms are intended to include the plural forms as well, unless the context clearly indicates otherwise. It will be further understood that the terms "comprising" and/or "including," when used in this specification, specify the presence of

stated features, wholes, steps, operations, elements, constituents and/or components, but do not preclude the presence or addition of one or more other features, wholes, steps, operations, elements, components, constituents and/or combinations thereof. It should be noted that the terms "up", "down", "front", "back", "left", "right", "inside", "outside" and similar expressions used herein are for illustrative purposes only and are not restrictive.

[0060] The exemplary embodiments of the present application will now be explained in further details with reference to the accompanying drawings.

[0061] The present application provides a silicon dioxide aerogel composite non-woven heat insulation sheet, comprising a non-woven and silicon dioxide aerogel. The non woven comprises a fiber and a binder bonding the fiber together, the binder having a hydroxyl group. The silicon dioxide aerogel is compounded on the non-woven. The silicon dioxide aerogel composite non-woven heat insulation sheet is obtained through a sol preparation process, a gel substrate composite process, a gel reaction process and a drying process.

[0062] Wherein, the sol preparation process includes heating reflux at a first preset time at a first preset temperature after a catalyst, solvent and silicon source are mixed and stirred evenly and adding and mixing evenly a gel accelerator to form a sol precursor after cooling. The gel substrate composite process includes compounding the sol precursor into the non-woven. The gel reaction process includes causing gel aging of a non-woven compounded with the sol precursor at a second preset temperature for a second preset time.

[0063] According to the silicon dioxide aerogel composite non-woven heat insulation sheet of the present application, the binder in the non-woven contains a hydroxyl group, which greatly reduces the powder loss rate of products.

[0064] As an implementation, the binder may contain polyvinyl alcohol, which is a compound with the hydroxyl group. Or, the binder may contain a reactant of polyacrylic acid and polyol. Or, the binder may contain polyvinyl alcohol and a reactant of polyacrylic acid and polyol, that is, the binder may be a mixture thereof.

[0065] Further preferably, the binder is polyvinyl alcohol. The non-woven is polyvinyl alcohol-based non-woven. Accordingly, the silicon dioxide aerogel composite non-woven heat insulation sheet of the present application may also be referred to as the silicon dioxide aerogel composite polyvinyl alcohol-based non-woven heat insulation sheet.

[0066] Wherein, PVA (polyvinyl alcohol) can be either pure polyvinyl alcohol or commercially available polyvinyl alcohol. For example, it may be hydrolyzed by polyvinyl acetate, which has the main chemical structure of polyvinyl alcohol but contains a small amount of polyvinyl acetate monomer or a small mixture of other monomers.

[0067] It is worth mentioning that the non-woven in the embodiment of the present application is a fiber felt formed without the need for spinning and weaving, i.e., non-woven fabric. Non-woven is formed by directionally or randomly arranging the short fiber or long fiber

to form a fiber network structure, and then reinforced by mechanical, thermal adhesion or chemical approaches, which does not have the warp and weft structure of textile fabric. Wherein, the fiber can choose glass fiber, carbon fiber, nylon fiber, polyester fiber, polyester fiber, polypropylene fiber or a mixture of several fibers.

[0068] The binder in the embodiment of this application refers to the binder used in non-woven fabrics to bring long and/or short fibers together or bind them together through physical and/or chemical action (e.g. intermolecular force) in three-dimensional space, i.e., the "binder", but not the "adhesive" used to form "surface to surface" bonding relationships between bonding surfaces.

[0069] The following will introduce the specific preparation method of the present application. Referring to FIG. 1, the silicon dioxide aerogel composite non-woven heat insulation sheet of this application is obtained successively through the sol preparation process, gel substrate composite process, gel reaction process, hydrophobic modification process, solvent replacement process and drying process.

[0070] In the sol preparation process, the catalyst is hydrochloric acid. The solvent comprises water and anhydrous ethanol. The silicon source is tetraethyl orthosilicate. The gel accelerator is ammonia water. The sol preparation process specifically includes adding hydrochloric acid and anhydrous ethanol after being mixed into tetraethyl orthosilicate and stirring for 2-5min, heating reflux mixed solution at 80-90°C for 3-5h, cooling to 15-25°C, and then adding ammonia water diluted with anhydrous ethanol and stirring for 15-25min to obtain the sol precursor.

[0071] The gel substrate composite process includes pouring the sol precursor into the non-woven by a perfusion process. Preferably, the gel substrate composite process can be a vacuum perfusion process.

[0072] The gel reaction process includes causing gel aging of a nonwoven compounded with the sol precursor for 20-28h at 30-40°C.

[0073] The hydrophobic modification process includes modifying a gel aged sheet with ethanol solution of silane of 20-30% volume fraction at 50-60°C for 8-24h.

[0074] The solvent replacement process includes treating a gel aged sheet with anhydrous ethanol at 50-60°C for 8-24h to replace all solvents with anhydrous ethanol.

[0075] The drying process is preferably CO_2 supercritical drying. Specifically, the drying process is performed using a CO_2 supercritical drying device with a drying kettle and a separating kettle.

[0076] Further, the drying process may comprise: the gel aged sheet is placed in the drying kettle and CO_2 is injected into the CO_2 supercritical drying device with a flow rate of 35-40 L/h for 6-10h, wherein the temperature in the drying kettle is controlled to be 50-70°C, and the pressure therein is controlled to be 17-18 MPa, so as to make CO_2 enter a supercritical

state and replace ethanol in the gel aged sheet; the temperature in the separating kettle is controlled to be 30-50°C and the pressure therein is controlled to be 7-8 MPa such that CO_2 and ethanol entering the separating kettle can be separated.

[0077] Exemplarily, the drying process can be carried out in the CO_2 supercritical drying device as shown in FIG. 2, wherein the CO_2 gas cylinder provides a gas source for the storage tank. The CO_2 gas forms liquid under the action of the refrigerator and enters the drying kettle with the booster pump. Preferably, in order to enable the sheet to be fully dried, a two-stage separating kettle is disposed for two separating steps.

[0078] The present application will be further explained in detail in combination with specific experimental procedures.

[0079] The non-woven substrate is cut to an appropriate size and stacked into the required thickness, for example, the sheet of 25cm×25cm can be cut and stacked into a thickness of 0.7cm~1cm. Different non-woven substrates can be taken as different embodiments in various embodiments. For example, the non-woven substrate can be W125 (which is produced by Owens Corning Company, with a gram weight of 125g/m^2, a single thickness of 1.11mm, binder containing PVA), YL-496 (which is produced by Owens Corning Company, with a gram weight of 59.5g/m^2, a single thickness of 0.5mm, binder containing PVA), CX-59 (which is produced by Owens Corning Company, with a gram weight of 46g/m^2, a single thickness of 0.4mm, binder containing PVA), Special NI (which is produced by Owens Corning Company, with a gram weight of 37g/m^2, a single thickness of 0.34mm, binder containing PVA), C50A (which is produced by Owens Corning Company, with a gram weight of 50g/m^2, a single thickness of 0.4mm, binder containing reactants of polyacrylic acid and polyol), and C30A (produced by Owens Corning Company, with a gram weight of 30g/m^2, a single thickness of 0.19mm, binder containing reactants of polyacrylic acid and polyol).

[0080] With 0.1mol/L of hydrochloric acid prepared, 43g of the diluted hydrochloric acid is mixed with 2450g of anhydrous ethanol in the reactor. 500g of tetraethyl orthosilicate is then added into the reactor and stirred for 3min with a magnetic agitator. After being mixed uniformly, the mixed solution is heated reflux at 85°C for 4h, wherein the temperature of the condenser tube is controlled at 5°C. The mixed solution is later cooled to 20°C.

[0081] 2200μL of 13mol/L ammonia water is diluted with 40g of anhydrous ethanol, and the diluted ammonia water is added to the mixed solution at 20°C and then stirred for 20min to obtain the sol precursor.

[0082] The sol precursor prepared by the step above is poured into the cut non-woven by vacuum perfusion process. The gel is then aged at 35°C for 24h. After that, the gel aged sheets is modified with ethanol solution of methyltrimethoxysilane of 20-30% volume fraction at 50-60°C for 8-24h. The gel aged and hydrophobic modified sheets is then treated with anhydrous ethanol at 50-60°C for 8-24h.

[0083] The gel aged sheet is successively placed in the drying kettle of the CO_2

supercritical drying device and CO_2 is injected into the CO_2 supercritical drying device with a flow rate of 35-40 L/h for 6-10h, wherein the temperature in the drying kettle is controlled to be 60°C, and the pressure therein is controlled to be 17-18 MPa. After all ethanol in the sheet is replaced with supercritical CO_2, the sheet is placed in the separating kettle. The temperature in the separating kettle is controlled to be 40°C, and the pressure therein is controlled to be 7.5MPa.

[0084] The present application will be further explained in detail in combination with Examples and Contrast Examples below.

[0085] In Example 1, W125 is selected as substrate, and the experiment of preparing the silicon dioxide aerogel composite non-woven heat insulation sheet is carried out according to the above specific steps.

[0086] In Example 2, YL-496 is selected as substrate, and the experiment of preparing the silicon dioxide aerogel composite non-woven heat insulation sheet is carried out according to the above specific steps.

[0087] In Example 3, CX-59 is selected as substrate, and the experiment of preparing the silicon dioxide aerogel composite non-woven heat insulation sheet is carried out according to the above specific steps.

[0088] In Example 4, Special NI is selected as substrate, and the experiment of preparing the silicon dioxide aerogel composite non-woven heat insulation sheet is carried out according to the above specific steps.

[0089] In Example 5, C50A is selected as substrate, and the experiment of preparing the silicon dioxide aerogel composite non-woven heat insulation sheet is carried out according to the above specific steps.

[0090] In Example 6, C30A is selected as substrate, and the experiment of preparing the silicon dioxide aerogel composite non-woven heat insulation sheet is carried out according to the above specific steps.

[0091] In Contract Example 1, C30A is selected as substrate, which is soaked in 0.1% PVA aqueous solution and then dried. The experiment of preparing the silicon dioxide aerogel composite non-woven heat insulation sheet is carried out according to the specific steps in the above embodiments.

[0092] In Contract Example 2, C30A is selected as substrate, which is soaked in 0.5% PVA aqueous solution and then dried. The experiment of preparing the silicon dioxide aerogel composite non-woven heat insulation sheet is carried out according to the specific steps in the above embodiments.

[0093] In Contract Example 3, C30A is selected as substrate, which is soaked in 1.0% PVA aqueous solution and then dried. The experiment of preparing the silicon dioxide aerogel composite non-woven heat insulation sheet is carried out according to the specific steps in the

above embodiments.

[0094] After the preparation of the silicon dioxide aerogel composite non-woven heat insulation sheet, it is weighed and put into a closed container, and then placed on the vibration equipment for shock, with the frequency of 1200rpm and the time of 10min. Upon the completion, it is weighted again, and the powder loss rate is calculated.

[0095] Refer to Table 1 for specific parameters and test results.

Table 1. Experimental comparison table of Examples 1-6 and Contrast Examples 1-3

Serial Number	Substrate	Binder	Treatment	Binder Content of Substrate	Load Rate (Aerogel / Non-woven)	Air Permeability (mm/s)	Powder Loss Rate (Weight Loss Ratio)	Heat Conductivity Coefficient (W/m • K)
Example 1	W125	Polyvinyl Alcohol	-	12%	32.1%	1340	0.46%	0.02082
Example 2	YL-496	Polyvinyl Alcohol	-	10.7%	31.1%	3018	0.39%	0.02045
Example 3	CX-59	Polyvinyl Alcohol	-	16.2%	-	1903	0.45%	-
Example 4	Special NI	Polyvinyl Alcohol	-	13%	-	5181	0.42%	0.02032
Example 5	C50A	Reactant of Polyacrylic Acid and Polyol	-	17%	35.9%	3823	0.4%	0.02079
Example 6	C30A	Reactant of Polyacrylic Acid and Polyol	-	10%	34.04%	5470	1.07%	0.01971
Contrast Example 1	C30A	Reactant of Polyacrylic Acid and Polyol	Being soaked in 0.1% PVA aqueous solution and then dried			-	2.17%	0.02115
Contrast Example 2	C30A	Reactant of Polyacrylic Acid and Polyol	Being soaked in 0.5% PVA aqueous solution and then dried			-	1.95%	0.01696
Contrast Example 3	C30A	Reactant of Polyacrylic Acid and Polyol	Being soaked in 1.0% PVA aqueous solution and then dried			-	1.4%	0.01849

[0096] Combined with the data of the above Examples 1-5, both PVA-based non-woven and polyacrylic acid-based non-woven can achieve better effect of reducing the powder loss rate.

[0097] Generally speaking, for non-woven, high air permeability means high porosity, and low air permeability means low porosity. In the actual usage experience of the silicon dioxide aerogel composite non-woven heat insulation sheet, the higher the porosity, the more

sparse the fiber of the non-woven, the weaker its ability to carry silicon dioxide aerogel particles, and the higher the powder loss rate.

[0098] Combined with the data of Examples 1-4 and 5-6, however, it can be seen that the PVA-based non-woven can also have better powder loss rate data under the condition that the air permeability changes. The powder loss rate of polyacrylic acid-based non-woven will be affected by air permeability, that is, the powder loss rate of C30A with high air permeability is significantly improved compared to that of C50A with low air permeability.

[0099] Apparently, PVA-based non-woven has a wider range of adaptability relative to the non-woven of other systems, and is free from the influence of air permeability. This makes the silicon dioxide aerogel composite non-woven heat insulation sheet prepared by various types of PVA-based non-woven can overcome the problem that the aerogel insulation sheet is easy to lose powder.

[00100] In combination with Example 6 and Contrast Examples 1-3, it can be seen that even if PVA adhesive is added to non-woven with non-PVA system, it cannot effectively reduce but increase the powder loss rate. Accordingly, when PVA serves as a skeleton binder to bind the fibers together in all directions of the space, it has a significant improvement in reducing the powder loss rate. However, when PVA serves as a surface adhesive, it does not significantly improve the powder loss rate.

[00101] In the silicon dioxide aerogel composite non-woven heat insulation sheet of the present application, the binder in the non-woven has a mass ratio of 5-30%, preferably 10%-17%. In the silicon dioxide aerogel composite non-woven heat insulation sheet, a ratio of the mass of the silicon dioxide aerogel to the mass of the non-woven is 5%-60%, preferably 32.1%-35.9%.

[00102] The silicon dioxide aerogel composite non-woven heat insulation sheet of the present application has an air permeability of 1000-6000 mm/s, preferably 1340-5470 mm/s. The silicon dioxide aerogel composite non-woven heat insulation sheet has a power loss rate of less than 1.5%, preferably less than 1.07%, more preferably less than 0.46%, and further preferably 0.39%-0.46%. The silicon dioxide aerogel composite non-woven heat insulation sheet has a heat conductivity coefficient of less than 0.021 W/m•K, preferably less than 0.02082 W/m•K, and more preferably 0.01971-0.02082 W/m•K. The silicon dioxide aerogel composite non-woven heat insulation sheet has a single sheet thickness of 0.3-3mm,. The silicon dioxide aerogel composite non-woven heat insulation sheet of the present application has an areal density of 30-300g/m^2.

[00103] The processes and steps described above in all preferred embodiments are examples only. Unless an adverse effect occurs, the various processing operations can be performed in a different order from the order of the above processes. The sequence of steps in the processes can also be added, combined, or subtracted according to actual needs.

[00104] Unless otherwise defined, the technical and scientific terms used herein have the

same meanings as commonly understood by those skilled in the technical field of the present application. The terms used herein are only for describing specific implementation purposes, and are not intended to limit the present application. A feature described in one embodiment herein can be applied to another embodiment alone or in combination with other features, unless the feature is not applicable in the other embodiment or otherwise stated.

[00105] The present application has been described through the above-mentioned embodiments, but it should be understood that the above-mentioned embodiments are only for the purpose of illustration and description. The present application is not limited to the above embodiments. More variations and modifications can be made according to the teachings of the present application, and these variations and modifications fall within the protection scope claimed by the present application.

WHAT IS CLAIMED IS:

1. A silicon dioxide aerogel composite non-woven heat insulation sheet, comprising:

a non-woven comprising a fiber and a binder bonding the fiber together, the binder having a hydroxyl group;

silicon dioxide aerogel compounded on the non-woven;

the silicon dioxide aerogel composite non-woven heat insulation sheet obtained through a sol preparation process, a gel substrate composite process, a gel reaction process and a drying process,

wherein the sol preparation process includes heating reflux at a first preset time at a first preset temperature after a catalyst, solvent and silicon source are mixed and stirred evenly and adding and mixing evenly a gel accelerator to form a sol precursor after cooling;

the gel substrate composite process includes compounding the sol precursor into the non-woven;

the gel reaction process includes causing gel aging of a non-woven compounded with the sol precursor at a second preset temperature for a second preset time.

2. The silicon dioxide aerogel composite non-woven heat insulation sheet of claim 1, wherein,

the binder contains polyvinyl alcohol; and/or

the binder includes a reactant of polyacrylic acid and polyol.

3. The silicon dioxide aerogel composite non-woven heat insulation sheet of claim 1, wherein,

the binder is polyvinyl alcohol.

4. The silicon dioxide aerogel composite non-woven heat insulation sheet of claim 1, wherein the catalyst is hydrochloric acid; the solvent comprises water and anhydrous ethanol; the silicon source is tetraethyl orthosilicate; and the gel accelerator is ammonia water.

5. The silicon dioxide aerogel composite non-woven heat insulation sheet of claim 4, wherein the sol preparation process includes adding hydrochloric acid and anhydrous ethanol after being mixed into tetraethyl orthosilicate and stirring for 2-5min, heating reflux mixed solution at 80-90°C for 3-5h, cooling to 15-25°C, and then adding ammonia water diluted with anhydrous ethanol and stirring for 15-25min to obtain the sol precursor.

6. The silicon dioxide aerogel composite non-woven heat insulation sheet of claim 1, wherein the gel reaction process includes causing gel aging of a nonwoven compounded with the sol precursor for 20-28h at 30-40°C.

7. The silicon dioxide aerogel composite non-woven heat insulation sheet of any one of claims 1-6, wherein a solvent replacement process is further included between the gel reaction process and the drying process, comprising:

treating a gel aged sheet with anhydrous ethanol at 50-60°C for 8-24h to replace all solvents with anhydrous ethanol.

8. The silicon dioxide aerogel composite non-woven heat insulation sheet of claim 7, wherein a hydrophobic modification process is further included between the gel reaction process and the solvent replacement process, comprising:

modifying a gel aged sheet with ethanol solution of silane of 20-30% volume fraction at 50-60°C for 8-24h.

9. The silicon dioxide aerogel composite non-woven heat insulation sheet of claim 7, wherein the drying process is CO_2 supercritical drying.

10. The silicon dioxide aerogel composite non-woven heat insulation sheet of claim 9, wherein the drying process is performed using a CO_2 supercritical drying device with a drying kettle and a separating kettle, comprising:

a gel aged sheet is placed in the drying kettle and CO_2 is injected into the CO_2 supercritical drying device with a flow rate of 35-40 L/h for 6-10h;

wherein the temperature in the drying kettle is controlled to be 50-70°C, and the pressure therein is controlled to be 17-18 MPa, so as to make CO_2 enter a supercritical state and replace ethanol in the gel aged sheet;

the temperature in the separating kettle is controlled to be 30-50°C and the pressure therein is controlled to be 7-8 MPa such that CO_2 and ethanol entering the separating kettle can be separated.

11. The silicon dioxide aerogel composite non-woven heat insulation sheet of any one of claims 1-6, wherein the gel substrate composite process includes pouring the sol precursor into the non-woven by a vacuum perfusion process.

12. The silicon dioxide aerogel composite non-woven heat insulation sheet of any one of claims 1-6, wherein the binder in the non-woven has a mass ratio of 5-30%.

13. The silicon dioxide aerogel composite non-woven heat insulation sheet of any one of claims 1-6, wherein in the silicon dioxide aerogel composite non-woven heat insulation sheet, a ratio of a mass of the silicon dioxide aerogel to a mass of the non-woven is 5%-60%.

14. The silicon dioxide aerogel composite non-woven heat insulation sheet of any one of claims 1-6, wherein the silicon dioxide aerogel composite non-woven heat insulation sheet has an air permeability of 1000-6000 mm/s.

15. The silicon dioxide aerogel composite non-woven heat insulation sheet of any one of claims 1-6, wherein the silicon dioxide aerogel composite non-woven heat insulation sheet has a power loss rate of less than 1.5%.

16. The silicon dioxide aerogel composite non-woven heat insulation sheet of any one of claims 1-6, wherein the silicon dioxide aerogel composite non-woven heat insulation sheet has a heat conductivity coefficient of less than 0.021 W/m•K.

17. The silicon dioxide aerogel composite non-woven heat insulation sheet of any one of claims 1-6, wherein the silicon dioxide aerogel composite non-woven heat insulation sheet has a thickness of 0.3-3mm.

18. The silicon dioxide aerogel composite non-woven heat insulation sheet of any one of claims 1-6, wherein the silicon dioxide aerogel composite non-woven heat insulation sheet has an areal density of 30-300g/m^2.

19. A silicon dioxide aerogel composite non-woven heat insulation sheet, comprising:

a non-woven comprising a fiber and a binder bonding the fiber together, the binder having a hydroxyl group;

silicon dioxide aerogel compounded on the non-woven.

20. The silicon dioxide aerogel composite non-woven heat insulation sheet of claim 19, wherein,

the binder contains polyvinyl alcohol; and/or

the binder includes a reactant of polyacrylic acid and polyol.

21. The silicon dioxide aerogel composite non-woven heat insulation sheet of claim 20, wherein,

the binder is polyvinyl alcohol.

22. The silicon dioxide aerogel composite non-woven heat insulation sheet of any one of claims 19-21, wherein the binder in the non-woven has a mass ratio of 5-30%.

23. The silicon dioxide aerogel composite non-woven heat insulation sheet of any one of claims 19-21, wherein in the silicon dioxide aerogel composite non-woven heat insulation sheet, a ratio of a mass of the silicon dioxide aerogel to a mass of the non-woven is 5%-60%.

24. The silicon dioxide aerogel composite non-woven heat insulation sheet of any one of claims 19-21, wherein the silicon dioxide aerogel composite non-woven heat insulation sheet has an air permeability of 1000-6000 mm/s.

25. The silicon dioxide aerogel composite non-woven heat insulation sheet of any one of claims 19-21, wherein the silicon dioxide aerogel composite non-woven heat insulation sheet has a power loss rate of less than 1.5%.

26. The silicon dioxide aerogel composite non-woven heat insulation sheet of any one of claims 19-21, wherein the silicon dioxide aerogel composite non-woven heat insulation sheet has a heat conductivity coefficient of less than 0.021 W/m•K.

27. The silicon dioxide aerogel composite non-woven heat insulation sheet of any one of claims 19-21, wherein the silicon dioxide aerogel composite non-woven heat insulation sheet has a thickness of 0.3-3mm.

28. The silicon dioxide aerogel composite non-woven heat insulation sheet of any one of claims 19-21, wherein the silicon dioxide aerogel composite non-woven heat insulation sheet has an areal density of 30-300g/m^2.

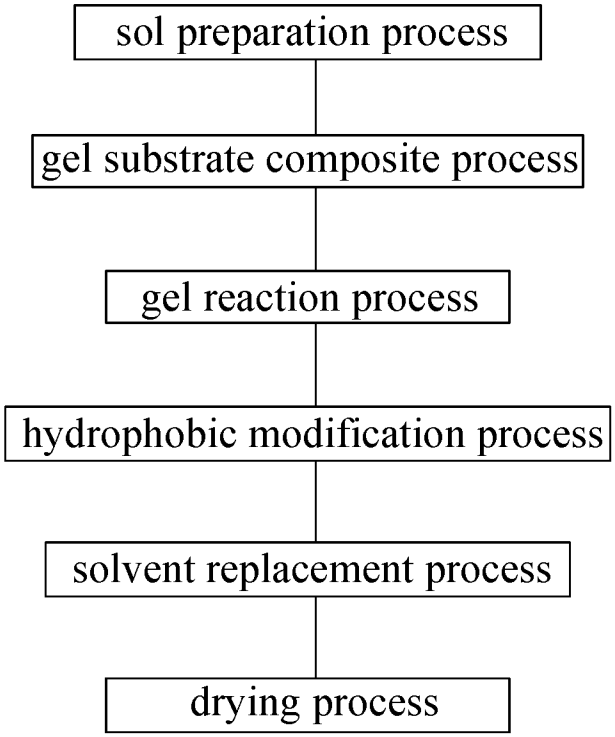
sol preparation process
gel substrate composite process
gel reaction process
hydrophobic modification process
solvent replacement process
drying process

FIG. 1

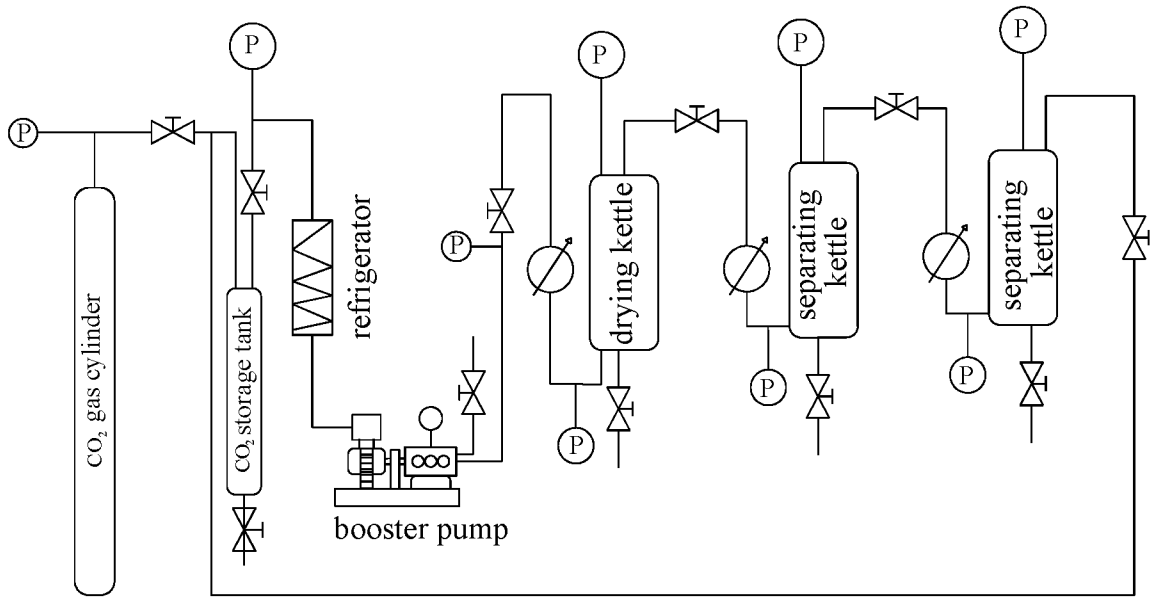
P
CO_2 gas cylinder
CO_2 storage tank
refrigerator
booster pump
drying kettle
separating kettle
separating kettle

FIG. 2

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2024/027929

A. CLASSIFICATION OF SUBJECT MATTER

INV. D04H1/413 B01J13/00 C01B33/154 C01B33/158 C04B14/06
D04H1/587 D04H1/64 D06M11/79 E04B1/76 E04B1/78
F16L59/02

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
D04H B01J C01B D06Q E04B F16L C04B D06M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2002/094426 A1 (STEPANIAN CHRISTOPHER J [US] ET AL) 18 July 2002 (2002-07-18) paragraph [0011] paragraph [0013] paragraph [0031] paragraph [0037] - paragraph [0040] paragraph [0049]; figures 1-2; examples 1, 2	1-28
X	US 2018/134566 A1 (KIM MI RI [KR] ET AL) 17 May 2018 (2018-05-17) paragraph [0094] - paragraph [0098]; figure 1; examples 1, 8	1-28
X	US 2019/145571 A1 (OIKAWA KAZUMA [JP] ET AL) 16 May 2019 (2019-05-16) paragraph [0025] - paragraph [0030]; examples 1, 4; table 1	1-28

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance;; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance;; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
11 July 2024	23/07/2024

Name and mailing address of the ISA/	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Demay, Stéphane

1

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/US2024/027929

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 2002094426 A1	18-07-2002	BR 0115523 A	16-09-2003
		CA 2429771 A1	04-07-2002
		CN 1592651 A	09-03-2005
		EP 1358373 A2	05-11-2003
		IL 155922 A	05-06-2008
		JP 6592050 B2	16-10-2019
		JP 2004517222 A	10-06-2004
		JP 2012182135 A	20-09-2012
		JP 2016001605 A	07-01-2016
		JP 2018012913 A	25-01-2018
		KR 20040030462 A	09-04-2004
		MX PA03004333 A	25-01-2005
		RU 2310702 C2	20-11-2007
		US 2002094426 A1	18-07-2002
		US 2006199455 A1	07-09-2006
		US 2009229032 A1	17-09-2009
		WO 02052086 A2	04-07-2002
US 2018134566 A1	17-05-2018	CN 107709235 A	16-02-2018
		CN 111501335 A	07-08-2020
		EP 3284721 A1	21-02-2018
		JP 6722283 B2	15-07-2020
		JP 2019501850 A	24-01-2019
		KR 20170104914 A	18-09-2017
		KR 20170104954 A	18-09-2017
		KR 20170104955 A	18-09-2017
		KR 20170104956 A	18-09-2017
		US 2018134566 A1	17-05-2018
US 2019145571 A1	16-05-2019	CN 109328280 A	12-02-2019
		EP 3480507 A1	08-05-2019
		JP 6667083 B2	18-03-2020
		JP 6967709 B2	17-11-2021
		JP 2020056504 A	09-04-2020
		JP WO2018003545 A1	11-04-2019
		US 2019145571 A1	16-05-2019
		WO 2018003545 A1	04-01-2018