

GRAPHENE INSOLE AND PREPARATION METHOD THEREFOR

TECHNICAL FIELD

The present application relates to the technical field of insoles, for example, a graphene insole and a preparation method therefor.

5 BACKGROUND

The quality of the insole is critical to the comfort level of footwear.

CN109700120A discloses a graphene antibacterial insole, which comprises a skin-contact layer, an antibacterial functional layer, and an elastic shock-absorption layer, wherein a fabric of the antibacterial functional layer contains graphene. The insole disclosed in CN109700120A has the
10 following beneficial effects. The disclosed antibacterial substance is graphene microsheets, and the graphene has an excellent antibacterial property due to its unique two-dimensional structure. The graphene is connected to chemical fiber macromolecules by chemical bonding, so that it is uniformly dispersed in the prepared chemical fiber, and then the resulting chemical fiber is woven into a fabric for insoles. The insole produced from such fabric has excellent antibacterial
15 activity, regardless of the presence of sweat, and meanwhile, because the graphene microsheets and the fabric macromolecules are connected by chemical bonds, the graphene microsheets are not easy to fall off or get losses, so that the prepared insole has a long antibacterial duration and washing resistance.

CN111518386A discloses a graphene foam and a production process therefor, and the disclosed
20 graphene foam is prepared from the following materials in mass percentages: 64.6-90.7% of polyurethane particles, 0.7-0.9% of water, 0.3-0.4% of a silicone oil foam-adjusting agent, 0.5-0.7% of a catalyst, 0.3-0.4% of an antibacterial agent, 5-30% of graphene, and 2.5-3% of special polyether polyol. Compared with the prior ordinary foam, the graphene foam disclosed in CN111518386A is prepared from a reasonable composition of the polyurethane particles, water,
25 silicone oil foam-adjusting agent, catalyst, antibacterial agent, graphene, and special polyether polyol by foaming process, and the graphene foam has more excellent air permeability, resilience, tear resistance, elongation, flexibility, thermal conductivity, antistatic property and other properties, which is especially suitable for insoles and mattresses.

At present, many researches on insoles mainly focus on the antimicrobial property of insoles,

while only few researches are conducted aiming at the functionality of insoles, and the cost of insole formulation with good functionality is high.

Therefore, developing an insole with excellent functionality and low cost is very important.

SUMMARY

- 5 The following is a summary of the subject described in detail herein. This summary is not intended to limit the protection scope of the claims.

10 An object of the present application is to provide a graphene insole and a preparation method therefor. The graphene insole can emit far-infrared radiation, which induces active hyperemia in blood vessels, boosts metabolism and improves blood fluidity, enhancing blood circulation in feet, thereby improving the human immune function, and the graphene insole can inhibit the growth of *Escherichia coli*, and has the properties of antibacterial activity, deodorization, antistatic function, heat preservation and ultraviolet resistance; the graphene insole also has excellent air permeability, water absorbency, and water desorption capability, which meet the requirements of foot protection and safety footwear; moreover, the cost of the insole is low.

- 15 To achieve the above object, the present application adopts the following technical solutions.

In a first aspect, examples of the present application provide a graphene insole, and raw materials for preparing the graphene insole comprise the following components in parts by weight:

	polyester polyol	24-30 parts;
	polyether polyol	20-26 parts;
20	diisocyanate	30-40 parts;
	glycerol	0.5-1.5 parts;
	other chain extender	1-7 parts;
	graphene	1-5 parts;
	water	1-3 parts;

silicone oil 0.1-2 parts;

amine additive 0.2-1 parts; and

diatomaceous earth 0.5-1.5 parts.

5 In examples of the present application, in terms of the raw materials for preparing the graphene
insole, the graphene is used in combination with glycerol, and also interacts with other
components in prescribed proportions, so that the formed graphene insole can emit far-infrared
radiation, which induces active hyperemia in blood vessels, boosts metabolism and improves
blood fluidity, enhancing blood circulation in feet, thereby improving the human immune
10 function, and the graphene insole can inhibit the growth of *Escherichia coli*, and has the
properties of antibacterial activity, deodorization, antistatic function, heat preservation and
ultraviolet resistance; moreover, the amount of graphene in the raw materials for preparing the
insole is small, and other raw materials are readily available, so the preparation of the graphene
insole is cost-efficient.

15 In an example of the present application, the polyester polyol is 24-30 parts by weight, such as
25 parts, 26 parts, 27 parts, 28 parts, 29 parts, etc.

The polyether polyol is 20-26 parts by weight, such as 21 parts, 22 parts, 23 parts, 24 parts, etc.

The diisocyanate is 30-40 parts by weight, such as 32 parts, 34 parts, 36 parts, 38 parts, etc.

The glycerol is 0.5-1.5 parts by weight, such as 0.6 parts, 0.8 parts, 1 part, 1.2 parts, 1.4 parts,
etc.

20 The other chain extender is 1-7 parts by weight, such as 2 parts, 3 parts, 4 parts, 5 parts, 6 parts,
etc.

The graphene is 1-5 parts by weight, such as 1.5 parts, 2 parts, 2.5 parts, 3 parts, 3.5 parts, 4 parts,
4.5 parts, 5 parts, etc.

25 The water is 1-3 parts by weight, such as 1.2 parts, 1.4 parts, 1.6 parts, 1.8 parts, 2 parts, 2.2
parts, 2.4 parts, 2.6 parts, 2.8 parts, etc.

The silicone oil is 0.1-2 parts by weight, such as 0.2 parts, 0.4 parts, 0.6 parts, 0.8 parts, 1 part,

1.2 parts, 1.4 parts, 1.6 parts, 1.8 parts, etc.

The amine additive is 0.2-1 parts by weight, such as 0.4 parts, 0.6 parts, 0.8 parts, etc.

The diatomaceous earth is 0.5-1.5 parts by weight, such as 0.6 parts, 0.8 parts, 1 part, 1.2 parts, 1.4 parts, etc.

- 5 In one example, the polyester polyol comprises dimer acid-based polyester polyol and/or phthalic anhydride-based polyester polyol.

In one example, a mass ratio of the dimer acid-based polyester polyol to the phthalic anhydride-based polyester polyol is 1: (1.5-3), wherein the “1.5-3” can be 1.6, 1.8, 2, 2.2, 2.4, 2.6, 2.8, etc.

- 10 In one example, the diisocyanate comprises diphenylmethane diisocyanate.

In one example, the other chain extender comprises ethylene glycol and/or diethanolamine, and further optionally a combination of ethylene glycol and diethanolamine.

In one example, the ethylene glycol is 2-5 parts by weight, such as 2.5 parts, 3 parts, 3.5 parts, 4 parts, 4.5 parts, etc.

- 15 In one example, the diethanolamine is 1-2 parts by weight, such as 1.2 parts, 1.4 parts, 1.6 parts, 1.8 parts, etc.

In one example, the silicone oil comprises open-cell silicone oil and/or closed-cell silicone oil, and further optionally a combination of open-cell silicone oil and closed-cell silicone oil.

- 20 In one example, the open-cell silicone oil is 0.5-1 parts by weight, such as 0.6 parts, 0.7 parts, 0.8 parts, 0.9 parts, etc.

In one example, the closed-cell silicone oil is 0.1-0.3 parts by weight, such as 0.12 parts, 0.14 parts, 0.16 parts, 0.18 parts, 0.2 parts, 0.22 parts, 0.24 parts, 0.26 parts, 0.28 parts, etc.

The raw materials further comprise polyphosphazene, and further optionally poly[bis(2,2,2-trifluoroethoxy)phosphazene].

- 25 In examples of the present application, due to the addition of polyphosphazene, the insole has

superior air permeability, water absorbency, and water desorption capability, which is suitable for foot protection and safety footwear, and it is ensured that the normal reflectivity and bacteriostasis of the insole are not affected adversely, and may be improved instead.

5 In one example, the polyphosphazene is 3-8 parts by weight, such as 4 parts, 5 parts, 6 parts, 7 parts, etc.

In one example, the raw materials further comprise a catalyst.

10 In one example, the catalyst comprises any one or a combination of at least two of dibutyltin dilaurate, pentamethyldiethylenetriamine, or an organic bismuth compound, wherein a typical but non-limiting combination comprises: a combination of dibutyltin dilaurate and pentamethyldiethylenetriamine, a combination of pentamethyldiethylenetriamine and an organic bismuth compound, and a combination of dibutyltin dilaurate, pentamethyldiethylenetriamine and an organic bismuth compound, etc.

In one example, the catalyst is 0.01-3 parts by weight, such as 0.05 parts, 0.1 parts, 0.5 parts, 1 part, 1.5 parts, 2 parts, 2.5 parts, etc.

15 As an optional technical solution, the raw materials for preparing the insole comprise the following components in parts by weight:

	polyester polyol	27 parts;
	polyether polyol	23 parts;
	diphenylmethane diisocyanate	36 parts;
20	glycerol	1 part;
	ethylene glycol	3.5 parts;
	diethanolamine	1.4 parts;
	graphene	3.5 parts;
	water	1.9 parts;

	open-cell silicone oil	0.8 parts;
	closed-cell silicone oil	0.2 parts;
	amine additive	0.6 parts;
	diatomaceous earth	1.1 parts;
5	polyphosphazene	5.5 parts; and
	catalyst	0.2 parts.

In a second aspect, the present application provides a preparation method for the graphene insole according to the first aspect, wherein the preparation method comprises the following steps:

10 (1) mixing polyester polyol, polyether polyol, glycerol, an other chain extender, graphene, water, an amine additive, diatomaceous earth, and silicone oil, and performing pre-polymerization to form a prepolymer; and

(2) mixing the prepolymer with diisocyanate, and performing polymerization and foaming and molding to obtain the graphene insole.

15 In one example, the pre-polymerization in step (1) is performed at a temperature of 50-100 °C, such as 55 °C, 60 °C, 65 °C, 70 °C, 75 °C, 80 °C, 85 °C, 90 °C, 95 °C, etc.

In one example, the pre-polymerization is performed for a period of 100-150 min, such as 110 min, 120 min, 130 min, 140 min, etc.

In one example, a manner of the mixing in step (2) comprises stirring.

20 In one example, the mixing is performed at a temperature of 20-22 °C, such as 20.2 °C, 20.4 °C, 20.6 °C, 20.8 °C, 21 °C, 21.2 °C, 21.4 °C, 21.6 °C, 21.8 °C, etc.

In one example, the polymerization is performed at a temperature of 100-140 °C, for example, 110 °C, 120 °C, 130 °C, etc.

In one example, the polymerization is performed for a period of 0.1-5 h, such as 0.5 h, 1 h, 1.5 h, 2 h, 2.5 h, 3 h, 3.5 h, 4 h, 4.5 h, etc.

As an optional technical solution, the preparation method comprises the following steps:

(1) mixing polyester polyol, polyether polyol, glycerol, an other chain extender, graphene, water, an amine additive, diatomaceous earth and silicone oil, and performing pre-polymerization at 50-100 °C for 100-150 min to form a prepolymer; and

- 5 (2) stirring to mix the prepolymer with diisocyanate at 20-22 °C, performing polymerization at 100-140 °C for 1-5 h, and performing foaming and molding to obtain the graphene insole.

Compared with the prior art, the present application has the following beneficial effects.

10 (1) In the present application, the graphene insole can emit far-infrared radiation, which induces active hyperemia in blood vessels, boosts metabolism and improves blood fluidity, enhancing blood circulation in feet, thereby improving the human immune function, and the graphene insole can inhibit the growth of *Escherichia coli*, and has the properties of antibacterial activity, deodorization, antistatic function, heat preservation and ultraviolet resistance.

15 (2) In the present application, the graphene insole also has excellent air permeability, water absorbency, and water desorption capability, which meet the requirements of foot protection and safety footwear.

(3) In the present application, the graphene insole has a far-infrared wavelength range of 4-16 μm , a normal reflectivity of more than or equal to 0.84, an air permeability of more than or equal to 109 mm/s, a water absorbency of more than or equal to 174 mg/cm², and a water desorption capability of more than or equal to 82%.

20 Other aspects will be appreciated upon reading and understanding the accompanying drawings and detailed description.

BRIEF DESCRIPTION OF DRAWINGS

25 Accompanying drawings are used to provide a further understanding of the technical solutions herein, form part of the specification, explain the technical solutions together with examples of the present application, and do not limit the technical solutions herein.

FIG. 1 is a test curve showing a far-infrared wavelength range of the graphene insole in Example 1.

DETAILED DESCRIPTION

In order to facilitate the understanding of the present application, examples of the present application are described below. Those skilled in the field should understand that the examples merely assist in understanding the present application and should not be regarded as a specific
5 limitation to the present application.

In the present application, purchase information of partial raw materials involved in each embodiment is as follows:

dimer acid-based polyester polyol is purchased from Shanghai Shawyten Technology Co., Ltd.;

10 phthalic anhydride-based polyester polyol is purchased from (Alfa) Zhengzhou Alfa Chemical Co., Ltd.;

polyether polyol is purchased from CNOOC and Shell Petrochemicals Co., Ltd., grade 2045;

an amine additive is purchased from Dongguan Chenrui New Materials Co., Ltd., grade AN130;

open-cell silicone oil is purchased from Momentive;

closed-cell silicone oil is purchased from Momentive;

15 diphenylmethane diisocyanate is purchased from Wanhua Chemical Group Co., Ltd, grade 8609E;

diatomaceous earth is purchased from Senda, grade 3005; and

polyphosphazene is purchased from Sigma-Aldrich (Shanghai) Trading Co., Ltd.

Example 1

20 This example provides a graphene insole, and raw materials for the graphene insole comprise the following components in parts by weight:

dimer acid-based polyester polyol	18 parts;
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phthalic anhydride-based polyester polyol	9 parts;
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	polyether polyol	23 parts;
	diphenylmethane diisocyanate	36 parts;
	glycerol	1 part;
	ethylene glycol	3.5 parts;
5	diethanolamine	1.4 parts;
	graphene	3.5 parts;
	water	1.9 parts;
	open-cell silicone oil	0.8 parts;
	closed-cell silicone oil	0.2 parts;
10	amine additive	0.6 parts;
	diatomaceous earth	1.1 parts; and
	dibutyltin dilaurate	0.2 parts.

The insole is prepared by the following method, and the method comprises the following steps:

15 (1) dimer acid-based polyester polyol, phthalic anhydride-based polyester polyol, polyether polyol, glycerol, ethylene glycol, diethanolamine, graphene, water, an amine additive, diatomaceous earth, dibutyltin dilaurate, open-cell silicone oil, and closed-cell silicone oil were mixed, and subjected to pre-polymerization at 80°C for 120 min to form a prepolymer; and

20 (2) then the prepolymer was pumped into a material storage cylinder A of a foaming machine, cooled to 21 °C by a refrigerating machine of the foaming machine, pumped together with diphenylmethane isocyanate into a mixing chamber by a transfer pump and stirred, then injected into a mold, polymerized at 120 °C for 30 min, and subjected to foaming and molding to obtain the graphene insole.

Example 2

This example provides a graphene insole, and raw materials for the graphene insole comprise the following components in parts by weight:

	dimer acid-based polyester polyol	9.6 parts;
	phthalic anhydride-based polyester polyol	14.4 parts;
5	polyether polyol	26 parts;
	diphenylmethane diisocyanate	30 parts;
	glycerol	0.5 parts;
	ethylene glycol	5 parts;
	diethanolamine	1 part;
10	graphene	1 part;
	water	3 parts;
	open-cell silicone oil	0.5 parts;
	closed-cell silicone oil	0.3 parts;
	amine additive	0.2 parts;
15	diatomaceous earth	1.5 parts;
	polyphosphazene	3 parts; and
	pentamethyldiethylenetriamine	0.5 parts.

The insole is prepared by the following method, and the method comprises the following steps:

- (1) dimer acid-based polyester polyol, phthalic anhydride-based polyester polyol, polyether polyol, glycerol, ethylene glycol, diethanolamine, graphene, water, an amine additive, diatomaceous earth, pentamethyldiethylenetriamine, open-cell silicone oil, and closed-cell silicone oil were mixed, and subjected to pre-polymerization at 100°C for 120 min to form a

prepolymer; and

- (2) then the prepolymer was pumped into a material storage cylinder A of a foaming machine, cooled to 20 °C by a refrigerating machine of the foaming machine, pumped together with polyphosphazene and diphenylmethane isocyanate into a mixing chamber by a transfer pump and stirred, then injected into a mold, polymerized at 100 °C for 30 min, and subjected to foaming and molding to obtain the graphene insole.

Example 3

This example provides a graphene insole, and raw materials for the graphene insole comprise the following components in parts by weight:

10	dimer acid-based polyester polyol	7.5 parts;
	phthalic anhydride-based polyester polyol	22.5 parts;
	polyether polyol	20 parts;
	diphenylmethane diisocyanate	40 parts;
	glycerol	1.5 parts;
15	ethylene glycol	2 parts;
	diethanolamine	2 parts;
	graphene	5 parts;
	water	1 part;
	open-cell silicone oil	1 part;
20	closed-cell silicone oil	0.1 parts;
	amine additive	1 part;
	diatomaceous earth	0.5 parts;

polyphosphazene 10 parts; and

dibutyltin dilaurate 1 part.

The insole is prepared by the following method, and the method comprises the following steps:

5 (1) dimer acid-based polyester polyol, phthalic anhydride-based polyester polyol, polyether polyol, glycerol, ethylene glycol, diethanolamine, graphene, water, an amine additive, diatomaceous earth, dibutyltin dilaurate, open-cell silicone oil, and closed-cell silicone oil were mixed, and subjected to pre-polymerization at 50°C for 120 min to form a prepolymer; and

10 (2) then the prepolymer was pumped into a material storage cylinder A of a foaming machine, cooled to 22 °C by a refrigerating machine of the foaming machine, pumped together with polyphosphazene and diphenylmethane isocyanate into a mixing chamber by a transfer pump and stirred, then injected into a mold, polymerized at 140 °C for 30 min, and subjected to foaming and molding to obtain the graphene insole.

Example 4

15 The difference between this example and Example 1 is that the open-cell silicone oil was replaced with closed-cell silicone oil with an equal mass, and the closed-cell silicone oil was 1 part by weight. The rest are the same as in Example 1.

Example 5

20 The difference between this example and Example 1 is that the closed-cell silicone oil was replaced with open-cell silicone oil with an equal mass, and the open-cell silicone oil was 1 part by weight. The rest are the same as in Example 1.

Example 6

The difference between this example and Example 1 is that the ethylene diamine was replaced with diethanolamine with an equal mass, and the diethanolamine was 4.9 parts by weight. The rest are the same as in Example 1.

Example 7

The difference between this example and Example 1 is that the diethanolamine was replaced

with ethylene diamine with an equal mass, and the ethylene diamine was 4.9 parts by weight. The rest are the same as in Example 1.

Example 8

- 5 The difference between this example and Example 1 is that 5.5 parts by weight of polyphosphazene was further comprised, which was mixed with the prepolymer in step (2). The rest are the same as in Example 1.

Comparative Example 1

The difference between this comparative example and Example 1 is that the glycerol was replaced with trimethylolpropane with an equal mass. The rest are the same as in Example 1.

10 Comparative Example 2

The difference between this comparative example and Example 1 is that the glycerol was not comprised, the ethylene glycol was 4.3 parts by weight, and the diethanolamine was 1.6 parts by weight. The rest are the same as in Example 1.

Comparative Example 3

- 15 The difference between this comparative example and Example 1 is that the dimer acid-based polyester polyol and the phthalic anhydride-based polyester polyol were replaced with polyether polyol with an equal mass, and the polyether polyol was 50 parts by weight. The rest are the same as in Example 1.

Comparative Example 4

- 20 The difference between this comparative example and Example 1 is that the polyether polyol was replaced with polyester polyol with an equal mass, the dimer acid-based polyester polyol was 33 parts by weight, and the phthalic anhydride-based polyester polyol was 17 parts by weight. The rest are the same as in Example 1.

Performance Test

- 25 The following tests are performed on the graphene insoles in Examples 1-8 and Comparative Examples 1-4:

(1) far-infrared wavelength range: the test was performed according to the standard of CAS 115-2005 Appendix A, and the wavelength was required to mainly distributed at 4-16 μm in the standard;

5 (2) normal reflectivity: the test was performed according to the standard of CAS 115-2005 Appendix A, and the normal reflectivity was required to be more than or equal to 0.80 in the standard;

(3) air permeability: the test was performed according to the standard of GB/T 5453-1997, an experimental area was 20 cm^2 , a pressure drop was 100 Pa, and the air permeability was required to be more than or equal to 100 mm/s;

10 (4) water absorbency: the test was performed according to the standard of GB/T 20991-2007, and the water absorbency was required to be more than or equal to 70 mg/cm^2 ; and

(5) water desorption capability: the test was performed according to the standard of GB/T 20991-2007, and the water desorption capability was required to be more than or equal to 80%.

The test results are summarized in Table 1.

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Table 1

	Far-infrared wavelength range (μm)	Normal reflectivity	Air permeability (mm/s)	Water absorbency (mg/cm^2)	Water desorption capability (%)
Standard requirement	4-16 μm	≥ 0.80	≥ 100 mm/s	≥ 70 mg/cm^2	$\geq 80\%$
Example 1	Qualified	0.89	121	178	88
Example 2	Qualified	0.88	123	280	90
Example 3	Qualified	0.87	134	285	91
Example 4	Qualified	0.86	115	174	82
Example 5	Qualified	0.87	109	175	83
Example 6	Qualified	0.88	112	176	84
Example 7	Qualified	0.84	114	177	86
Example 8	Qualified	0.90	137	295	100
Comparative Example 1	Unqualified	--	105	170	80
Comparative Example 2	Unqualified	--	102	165	76
Comparative	Unqualified	--	95	72	71

Example 3					
Comparative Example 4	Unqualified	--	91	64	65

In the table, "--" represents that the test object does not meet the standard and the data is not provided.

By the analysis of the data in Table 1, it can be seen that in the present application, the graphene insole can emit far-infrared radiation, which induces active hyperemia in blood vessels, boosts metabolism and improves blood fluidity, enhancing blood circulation in feet, thereby improving the human immune function, and the graphene insole also can inhibit the growth of *Escherichia coli*, and has the properties of antibacterial activity, deodorization, antistatic function, heat preservation and ultraviolet resistance; the graphene insole in the present application has the far-infrared wavelength range of 4-16 μm , and the normal reflectivity of more than or equal to 0.84, which meet the standard requirements of CAS 115-2005 "Health-care Textiles"; exemplarily, FIG. 1 is a test curve showing a far-infrared wavelength range of the graphene insole in Example 1, wherein the relative radiation intensity is obtained by normalization based on the maximum value; moreover, the graphene insole in the present application has excellent air permeability of more than or equal to 109 mm/s, excellent water absorbency of more than or equal to 174 mg/cm², and excellent water desorption capability of more than or equal to 82%.

By the analysis of Comparative Examples 1-2 and Example 1, it can be seen that the performance of the insole in Comparative Examples 1-2 is inferior to that in Example 1, thus proving the addition of glycerol is conducive to improving the performance of the insole in the present application.

By the analysis of Comparative Examples 3-4 and Example 1, it can be seen that the performance of the insole in Comparative Examples 3-4 is inferior to that in Example 1, thus proving the combined use of polyester polyol and polyether polyol is conducive to improving the performance of the insole in the present application.

By the analysis of Examples 4-5 and Example 1, it can be seen that the performance of the insole in Examples 4-5 is inferior to that in Example 1, thus proving the combined use of open-cell silicone oil and closed-cell silicone oil is conducive to improving the performance of the insole in the present application.

By the analysis of Examples 6-7 and Example 1, it can be seen that the performance of the insole in Examples 6-7 is inferior to that in Example 1, thus proving the combined use of ethylene

glycol and diethanolamine is conducive to improving the performance of the insole in the present application.

By the analysis of Example 8 and Example 1, it can be seen that the performance of the insole in Example 8 is inferior to that in Example 1, thus proving the addition of polyphosphazene endows the insole with better performance.

The applicant declares that the detailed solutions in the present application are illustrated by the above examples, but the present application is not limited to the above-detailed solutions, that is, the present application does not necessarily rely on the above-detailed solutions to be implemented. Those skilled in the art should understand that any improvement of the present application, the equivalent substitution of each raw material of products, the addition of auxiliary ingredients, and the selection of specific methods in the present application shall fall within the protection scope and disclosure scope of the present application.

What is claimed is:

1. A graphene insole, which is prepared from raw materials comprising the following components in parts by weight:

	polyester polyol	24-30 parts;
5	polyether polyol	20-26 parts;
	diisocyanate	30-40 parts;
	glycerol	0.5-1.5 parts;
	other chain extender	1-7 parts;
	graphene	1-5 parts;
10	water	1-3 parts;
	silicone oil	0.1-2 parts;
	amine additive	0.2-1 parts; and
	diatomaceous earth	0.5-1.5 parts.

15 2. The graphene insole according to claim 1, wherein the polyester polyol comprises dimer acid-based polyester polyol and/or phthalic anhydride-based polyester polyol.

3. The graphene insole according to claim 2, wherein a mass ratio of the dimer acid-based polyester polyol to the phthalic anhydride-based polyester polyol is 1: (1.5-3).

4. The graphene insole according to any one of claims 1-3, wherein the diisocyanate comprises diphenylmethane diisocyanate.

20 5. The graphene insole according to any one of claims 1-4, wherein the other chain extender comprises ethylene glycol and/or diethanolamine.

6. The graphene insole according to claim 5, wherein the ethylene glycol is 2-5 parts by weight;

optionally, the diethanolamine is 1-2 parts by weight.

7. The graphene insole according to any one of claims 1-6, wherein the silicone oil comprises open-cell silicone oil and/or closed-cell silicone oil;

optionally, the open-cell silicone oil is 0.5-1 parts by weight;

5 optionally, the closed-cell silicone oil is 0.1-0.3 parts by weight.

8. The graphene insole according to any one of claims 1-7, wherein the raw materials further comprise polyphosphazene;

optionally, the polyphosphazene is 3-8 parts by weight;

optionally, the raw materials further comprise a catalyst.

10 9. The graphene insole according to claim 8, wherein the catalyst comprises any one or a combination of at least two of dibutyltin dilaurate, pentamethyldiethylenetriamine, or an organic bismuth compound;

optionally, the catalyst is 0.01-3 parts by weight.

15 10. A preparation method for the graphene insole according to any one of claims 1-9, comprising the following steps:

(1) mixing polyester polyol, polyether polyol, glycerol, an other chain extender, graphene, water, an amine additive, diatomaceous earth, and silicone oil, and performing pre-polymerization to form a prepolymer; and

20 (2) mixing the prepolymer with diisocyanate, and performing polymerization and foaming and molding to obtain the graphene insole.

11. The preparation method according to claim 10, wherein the pre-polymerization in step (1) is performed at a temperature of 50-100 °C.

12. The preparation method according to claim 10 or 11, wherein the pre-polymerization is performed for a period of 100-150 min.

13. The preparation method according to any one of claims 10-12, wherein a manner of the mixing in step (2) comprises stirring.
14. The preparation method according to any one of claims 10-13, wherein the mixing is performed at a temperature of 20-22 °C.
- 5 15. The preparation method according to any one of claims 10-14, wherein the polymerization is performed at a temperature of 100-140 °C.
16. The preparation method according to any one of claims 10-15, wherein the polymerization is performed for a period of 0.1-5 h.
17. The preparation method according to any one of claims 10-16, wherein the preparation
10 method comprises the following steps:
- (1) mixing polyester polyol, polyether polyol, glycerol, an other chain extender, graphene, water, an amine additive, diatomaceous earth and silicone oil, and performing pre-polymerization at 50-100 °C for 100-150 min to form a prepolymer; and
- (2) stirring to mix the prepolymer with diisocyanate at 20-22 °C, performing polymerization at
15 100-140 °C for 1-5 h, and performing foaming and molding to obtain the graphene insole.