FORM 2

THE PATENT ACT, 1970

(39 of 1970)

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The Patent Rules, 2003

COMPLETE SPECIFICATION

(See section 10 and rule 13)

1. TITLE OF THE INVENTION:

A Method for Synthesis of Bio-Sourced Epoxy Vitrimer for Enhanced Toughness and Recyclability

2. APPLICANT:

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3. PREAMBLE TO THE DESCRIPTION:

The following specification particularly describes the invention and the manner in which it is to be performed:

4. DESCRIPTION:

Field of the invention:

[0001] The present disclosure generally relates to the technical field of materials science, and in specific relates to a method for developing a bio-sourced epoxy vitrimer with enhanced toughness, recyclability, and stress relaxation properties, while partially replacing petrochemical-based components with bio-based alternatives to reduce environmental impact and promote sustainable material solutions.

Background of the invention:

[0002] In recent year, driven by global concerns over resource depletion and environmental degradation, the need for sustainable and environmentally friendly materials has significantly increased. Among various materials, epoxies are widely used in industries such as automotive, aerospace, and electronics due to their excellent mechanical properties, thermal stability, and chemical resistance. However, conventional epoxies are predominantly derived from petrochemical sources, raising concerns about their environmental impact, particularly in terms of recyclability and the carbon footprint associated with their production.

[0003] However, the production of traditional epoxy resins relies heavily on petrochemical resources, contributing to environmental pollution and resource depletion. Additionally, these materials are not easily recyclable, leading to waste management issues. Further, the thermosetting nature of conventional epoxies makes them difficult to recycle or reprocess, as they form irreversible crosslinked networks upon curing. This results in a significant accumulation of non-recyclable waste. Despite their widespread use, conventional epoxies often suffer from brittleness, which limits their toughness and makes them prone to cracking under stress. This mechanical limitation restricts their applicability in demanding environments.

[0004] Efforts have been made to replace petrochemical-based components with bio-based alternatives, such as epoxies derived from natural sources like vegetable oils or lignin. These bio-based epoxies aim to reduce the environmental impact of epoxy production. Although bio-based epoxies reduce reliance on petrochemical resources, they often struggle to match the mechanical performance of their petrochemical counterparts. Issues such as lower toughness and reduced thermal stability can limit their use in high-performance applications.

[0005] To overcome the mechanical limitations of conventional epoxies, fiber-reinforced composite materials have been developed. These composites combine epoxy resins with reinforcing fibers (e.g., carbon or glass) to enhance toughness and strength. While effective in improving toughness, fiber-reinforced composites are still dependent on petrochemical-based epoxy matrices. Additionally, the recycling of these composites is complex due to the difficulty in separating the fibers from the resin matrix, leading to limited practical recyclability.

[0006] A newer class of materials, known as vitrimers, has been developed to introduce dynamic covalent bonds within the epoxy network. These bonds allow for the reprocessing and recycling of the material under certain conditions, thereby improving its recyclability. The development of vitrimers has opened new possibilities for recyclable thermoset resins, but many existing formulations rely on petrochemical-based epoxies. This limits their environmental benefits and does not fully address the need for sustainable material sources. Moreover, achieving the desired balance of mechanical properties and reprocessability remains challenging.

[0007] Hence, the existing thermoset resins face significant challenges in terms of environmental sustainability. The existing thermoset resins is made of an irreversible network structure, which makes them unsuitable for thermal reprocessing, unlike thermoplastics. Although this cross-linked structure provides enhanced chemical resistance, it also creates a major barrier to depolymerizing thermosets and recovering their basic components for re-polymerization.

[0008] Therefore, there is a need for a method for developing a bio-sourced epoxy vitrimer with enhanced toughness, recyclability, and stress relaxation properties. There is also a need for a bio-sourced epoxy vitrimer that is developed by partially or completely replacing petrochemical-based components with bio-based alternatives to reduce environmental impact and promote sustainable material solutions. There is also a need for a bio-sourced epoxy vitrimer that exhibits ability to be reprocessed and recycled multiple times without significant degradation of material properties.

Objectives of the invention:

[0009] The primary objective of the invention is to provide a method for developing a bio-sourced epoxy vitrimer with enhanced toughness, recyclability, and stress relaxation properties.

[0010] The other objective of the invention is to provide a bio-sourced epoxy vitrimer that can combine thermal re-processability of thermoplastics with the robust network structure of thermosets.

[0011] Another objective of the invention is to develop a bio-sourced epoxy vitrimer that is developed by partially or completely replacing petrochemical-based components with bio-based alternatives to reduce environmental impact and promote sustainable material solutions.

[0012] The other objective of the invention is to develop a bio-sourced epoxy vitrimer that exhibits ability to be reprocessed and recycled multiple times without significant degradation of material properties.

[0013] Another objective of the invention is to develop a bio-sourced epoxy vitrimer with enhanced toughness and flexibility compared to traditional epoxies, broadening potential applications.

[0014] Another objective of the invention is to provide a bio-sourced epoxy vitrimer that provides lower carbon footprint and reduced resource depletion.

[0015] The other objective of the invention is to provide a bio-sourced epoxy vitrimer with potential for use in various applications, including automotive components, aerospace, and electronics, due to customizable properties.

[0016] The other objective of the invention is to provide method for developing a bio-sourced epoxy vitrimer at reduced production costs through the use of bio-based materials and efficient recycling processes.

Summary of the invention:

[0017] The present disclosure proposes a method for synthesis of bio-sourced epoxy vitrimer for enhanced toughness and recyclability. The following presents a simplified summary in order to provide a basic understanding of some aspects of the claimed subject matter. This summary is not an extensive overview. It is not intended to identify key/critical elements or to delineate the scope of the claimed subject matter. Its sole purpose is to present some concepts in a simplified form as a prelude to the more detailed description that is presented later.

[0018] In order to overcome the above deficiencies of the prior art, the present disclosure is to solve the technical problem to provide a method for developing a bio-sourced epoxy vitrimer with enhanced toughness, recyclability, and stress relaxation properties, while partially replacing petrochemical-based components with bio-based alternatives to reduce environmental impact and promote sustainable material solutions.

[0019] According to an aspect, the invention provides a bio-sourced epoxy vitrimer that comprises 70 to 75 weight percentage of an imine hardener and 25 to 30 weight percentage of a bio-epoxy blend. In one embodiment, the imine hardener is synthesized by mixing a vanillin solution with a hardening agent in a molar ratio of 1:2.

[0020] In one embodiment, the vanillin solution is prepared by mixing 0.25 moles of vanillin in 100 mL of methanol. The hardening agent is 3, 3'-dimethyl-4, 4'-diaminodicyclohexylmethane (DMDC). The vanillin solution is mixed with 0.5 moles of DMDC.

[0021] The bio-epoxy blend comprises epoxy methyl ricinoleate (EMR) and an epoxy resin. In one embodiment, the EMR is blended with the epoxy resin in varying proportions to partially replace petro-based epoxy components, achieving an optimal balance between stiffness and toughness.

[0022] According to another aspect, the invention provides a method for preparing the bio-sourced epoxy vitrimer. At first, the the vanillin solution is mixed with the hardening agent under an inert atmosphere and at atmospheric pressure to form a reaction mixture. Next, the reaction mixture is heated to a temperature within a range of 50°C to 70°C and continuously stirred for a time period of 3 to 6 hr. Subsequently, the reaction mixture is cooled down to ambient temperature and excess DMDC is removed from the reaction mixture to a precipitate.

[0023] Next, the precipitate is rinsed with methanol, and dried under vacuum at a temperature ranging from 80 °C to 120 °C for a time period of 12 to 24 hr to obtain the imine hardener. Next, a mixture of epoxy methyl ricinoleate (EMR) and the epoxy resin is subjected through stirring and ultrasonication for a time period of 30 min to obtain the bio-epoxy blend. In a preferred embodiment, the EMR is mixed with the epoxy resin in various weight ratios, specifically 10%, 20%, and 30% of obtain different bio-epoxy blends.

[0024] Next, the imine hardener is blended with the bio-epoxy blend to obtain an epoxy mixture. In one embodiment, the imine hardener is blended with the bio-epoxy blend at a ratio of 3:1. Later, the epoxy mixture is curried to form the bio-sourced epoxy vitrimer with enhanced toughness, recyclability, and stress relaxation properties. The epoxy mixture is cured sequentially at different temperatures: first at 100 °C for a time period of 2 hr, then at 130 °C for a time period of 2 hr, and finally at 150 °C for a time period of 2 hr.

[0025] Further, objects and advantages of the present invention will be apparent from a study of the following portion of the specification, the claims, and the attached drawings.

Detailed description of drawings:

[0026] The accompanying drawings, which are incorporated in and constitute a part of the specification, illustrate an embodiment of the invention, and, together with the description, explain the principles of the invention.

[0027] FIG. 1 illustrates a flowchart of a method for preparing a bio-sourced epoxy vitrimer, in accordance to an exemplary embodiment of the invention.

[0028] FIG. 2 illustrates a scheme for synthesis of an imine hardener, in accordance to an exemplary embodiment of the invention.

Detailed invention disclosure:

[0029] Various embodiments of the present invention will be described in reference to the accompanying drawings. Wherever possible, same or similar reference numerals are used in the drawings and the description to refer to the same or like parts or steps.

[0030] The present disclosure has been made with a view towards solving the problem with the prior art described above, and it is an object of the present invention to provide a method for developing a bio-sourced epoxy vitrimer with enhanced toughness, recyclability, and stress relaxation properties, while partially replacing petrochemical-based components with bio-based alternatives to reduce environmental impact and promote sustainable material solutions. The bio-sourced epoxy vitrimer, obtained from at least 50 weight percentage of bio-source as raw material. The bio-sourced epoxy vitrimer exhibits enhanced toughness, recyclability, and stress relaxation properties, and is capable of being reprocessed and recycled under moderate thermal conditions.

[0031] According to an exemplary embodiment of the invention, FIG. 1 refers to a flowchart 100 of a method for preparing a bio-sourced epoxy vitrimer. The bio-sourced epoxy vitrimer comprises 70 to 75 weight percentage of an imine hardener and 25 to 30 weight percentage of a bio-epoxy blend. In one embodiment, the imine hardener is synthesized by mixing a vanillin solution with a hardening agent in a molar ratio of 1:2.

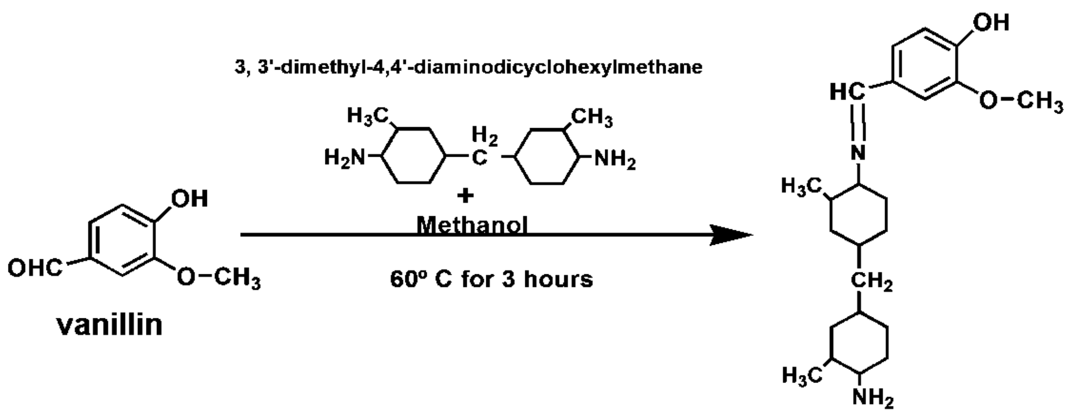
[0032] In one embodiment, the vanillin solution is prepared by mixing 0.25 moles of vanillin in 100 mL of methanol. The hardening agent is 3, 3'-dimethyl-4, 4'-diaminodicyclohexylmethane (DMDC). The vanillin solution is mixed with 0.5 moles of DMDC in a three-necked round-bottomed flask featuring a reflux condenser. The bio-epoxy blend comprises epoxy methyl ricinoleate (EMR) and an epoxy resin. In one embodiment, the EMR is blended with the epoxy resin in varying proportions to partially replace petro-based epoxy components, achieving an optimal balance between stiffness and toughness. The imine hardener is introduced to the bio-epoxy blend in a 3:1 ratio. Therefore, in a preferred embodiment, the bio-sourced epoxy vitrimer comprises 75 weight percentage of the imine hardener and 25 weight percentage of the bio-epoxy blend.

[0033] Referring to FIG. 1, the vanillin solution is mixed with the hardening agent under an inert atmosphere and at atmospheric pressure to form a reaction mixture, as depicted in step 102. At step 104, the reaction mixture is heated to a temperature within a range of 50 °C to 70 °C and continuously stirred for a time period of 3 to 6 hr. In a preferred embodiment, the reaction mixture is heated to a temperature 60 °C and stirred continuously for a time period of 5 hr. Subsequently, the reaction mixture is cooled down to ambient temperature and excess DMDC is removed from the reaction mixture to a precipitate. The precipitate obtain is a yellow coloured precipitate.

[0034] At step 106, the precipitate is rinsed with methanol, and dried under vacuum at a temperature ranging from 80 °C to 120 °C for a time period of 12 to 24 hr to obtain the imine hardener. In a preferred embodiment, after rinsing, the precipitate is dried under vacuum at a temperature of 100 °C for a time period of 24 hr to obtain the imine hardener.

[0035] FIG. 2 illustrates a scheme 200 for synthesis of the imine hardener, and is presented in Scheme 1.

Scheme 1



[0036] At step 108, a mixture of epoxy methyl ricinoleate (EMR) and the epoxy resin is subjected through stirring and ultrasonication for a time period of 30 min to obtain the bio-epoxy blend. In a preferred embodiment, the EMR is mixed with the epoxy resin in various weight ratios, specifically 10%, 20%, and 30% of obtain different bio-epoxy blends. These ratios of bio-epoxy blends are designated as EPEMR10, EPEMR20, and EPEMR30, respectively. Each bio-epoxy blend is mechanically stirred and ultrasonicated for 30 min to ensure thorough mixing. Following this, each bio-epoxy blend is placed in a vacuum oven to eliminate any air bubbles and then cooled to room temperature.

[0037] At step 110, the imine hardener is blended with each bio-epoxy blend to obtain an epoxy mixture. In one embodiment, the imine hardener is blended with the bio-epoxy blend at a ratio of 3:1. At step 112, the epoxy mixture is curried to form the bio-sourced epoxy vitrimer with enhanced toughness, recyclability, and stress relaxation properties. The epoxy mixture is cured sequentially at different temperatures: first at 100 °C for a time period of 2 hr, then at 130 °C for a time period of 2 hr, and finally at 150 °C for a time period of 2 hr.

[0038] In one embodiment, the bio-epoxy blend is incorporated into the bio-sourced epoxy vitrimer in proportions ranging from 10% to 50% by weight, to replace a corresponding portion of petro-based epoxy resin. In a preferred embodiment, EPEMR20 is effective in enhancing properties of the bio-sourced epoxy vitrimer.

[0039] In one embodiment, the bio-sourced epoxy vitrimer is characterized by enhanced toughness, recyclability, and stress relaxation properties, and is capable of being reprocessed and recycled under moderate thermal conditions. The bio-sourced epoxy vitrimer exhibits a relaxation curve with a decrease in duration at increasing temperatures, attributed to the active exchange reaction of Schiff base at elevated temperatures.

[0040] In one embodiment, the bio-sourced epoxy vitrimer is combined with natural fibers, such as flax fibers to form a fiber-reinforced composite material. The fiber-reinforced composite material exhibits improved environmental sustainability and mechanical properties. The bio-sourced epoxy vitrimer's matrix provides enhanced reprocessability and recyclability under moderate thermal conditions, while maintaining optimal mechanical strength and toughness. The use of the bio-sourced epoxy vitrimer as a replacement for petro-based epoxy resins in the production of composite materials for automotive interior parts.

[0041] According to another exemplary embodiment of the invention, a study is conducted on the synthesized bio-sourced epoxy vitrimer. The study further examines the relaxation behavior of the synthesized bio-sourced epoxy vitrimer, where the relaxation curve of EPEMR20-V demonstrates a reduction in relaxation duration with increasing temperature from 28.42 min at 80°C to 1.71 min at 120 °C. This behaviour is attributed to the active exchange reaction of Schiff base at elevated temperatures, which accelerates the reorganization of the network topology and, consequently, increases the rate of relaxation. Additionally, the invention includes the formulation and characterization of composites comprising vitrimers and flax fibers, enhancing their environmental sustainability.

[0042] In one embodiment, the synthesized bio-sourced epoxy vitrimer combines the thermal re-processability of thermoplastics with the robust network structure of thermosets. In the synthesized bio-sourced epoxy vitrimer, dynamic covalent bonds are integrated into thermosetting materials, leading to the creation of Covalently Adaptable Networks (CANs). These networks, while being strongly cross-linked, possess the ability to undergo stress relaxation and reversible de-polymerization through the breaking and reforming/exchanging of crosslinks. Additionally, the current invention addresses the issue of reducing reliance on petro-based epoxies by incorporating bio-epoxy, which also enhances toughness, recyclability, and stress relaxation.

[0043] In another embodiment, automotive industries use a vast array of plastic components, with over 30,000 parts in a vehicle, approximately one-third of which are made from plastics. The majority of these plastics are derived from non-biodegradable, petroleum-based sources, such as polypropylene, polyurethane, polyamides, and PVC, all of which have significant environmental impacts. The introduction of bio-resins and their natural fiber-reinforced composite systems offers a promising alternative to these petro-based materials. The synthesized bio-sourced epoxy vitrimer and their composites could replace the interior parts of automobiles currently made from petro-based polymers, providing a more sustainable and environmentally friendly option that meets industry standards and performance requirements.

[0044] Numerous advantages of the present disclosure may be apparent from the discussion above. In accordance with the present disclosure, a method is disclosed here for developing a bio-sourced epoxy vitrimer with enhanced toughness, recyclability, and stress relaxation properties.

[0045] The bio-sourced epoxy vitrimer combines thermal re-processability of thermoplastics with the robust network structure of thermosets. The bio-sourced epoxy vitrimer is developed by partially or completely replacing petrochemical-based components with bio-based alternatives to reduce environmental impact and promote sustainable material solutions.

[0046] The bio-sourced epoxy vitrimer exhibits ability to be reprocessed and recycled multiple times without significant degradation of material properties. The bio-sourced epoxy vitrimer is configured with enhanced toughness and flexibility compared to traditional epoxies, broadening potential applications. The bio-sourced epoxy vitrimer provides lower carbon footprint and reduced resource depletion. The bio-sourced epoxy vitrimer is used in various applications, including automotive components, aerospace, and electronics, due to customizable properties.

[0047] The proposed method develops the bio-sourced epoxy vitrimer at reduced production costs through the use of bio-based materials and efficient recycling processes. Thereby, making the bio-sourced epoxy vitrimer cost effective.

[0048] It will readily be apparent that numerous modifications and alterations can be made to the processes described in the foregoing examples without departing from the principles underlying the invention, and all such modifications and alterations are intended to be embraced by this application.

5. CLAIMS:

I / We Claim:

1. A bio-sourced epoxy vitrimer, comprising:

   70 to 75 weight percentage of an imine hardener; and

   25 to 30 weight percentage of a bio-epoxy blend,

   wherein the bio-sourced epoxy vitrimer exhibits enhanced toughness, recyclability, and stress relaxation properties, and is capable of being reprocessed and recycled under moderate thermal conditions.

2. The bio-sourced epoxy vitrimer as claimed in claim 1, wherein the imine hardener is synthesized by mixing a vanillin solution with a hardening agent.

3. The bio-sourced epoxy vitrimer as claimed in claim 2, wherein the vanillin solution comprises at least 0.25 moles of vanillin in methanol, and the hardening agent comprises at least 0.5 moles of 3,3'-dimethyl-4,4'-diaminodicyclohexylmethane (DMDC).

4. The bio-sourced epoxy vitrimer as claimed in claim 1, wherein the bio-epoxy blend comprises epoxy methyl ricinoleate (EMR) and an epoxy resin.

5. A method for preparing a bio-sourced epoxy vitrimer, comprising

   mixing a vanillin solution with a hardening agent under an inert atmosphere and at atmospheric pressure to form a reaction mixture;

   heating the reaction mixture to a temperature within a range of 50°C to 70°C and maintaining stirring for a time period of 3 to 6 hr, followed by cooling the reaction mixture at an ambient temperature for obtaining a precipitate;

   rinsing the precipitate with a solvent and drying the precipitate under vacuum at a temperature varies between 80 °C and 120 °C for a time period of at least 12 to 24 hr to obtain an imine hardener;

   subjecting a mixture of epoxy methyl ricinoleate (EMR) and an epoxy resin through stirring and ultrasonication for a time period of at least 30 min to obtain a bio-epoxy blend;

blending and heating the imine hardener with the bio-epoxy blend to obtain an epoxy mixture; and

curing the epoxy mixture to form the bio-sourced epoxy vitrimer that exhibits enhanced toughness, recyclability, and stress relaxation properties.

6. The method as claimed in claim 5, wherein the solvent is methanol.

7. The method as claimed in claim 5, wherein the imine hardener is blended with the bio-epoxy blend at a ratio of 3:1.

8. The method as claimed in claim 5, wherein the diamine hardening agent comprises 3,3'-dimethyl-4,4'-diaminodicyclohexylmethane (DMDC) for forming imine bond.

9. The method as claimed in claim 5, wherein the EMR is blended with the epoxy resin in varying proportions ranging from 10% to 30%.

10. The method as claimed in claim 5, wherein the epoxy mixture is cured sequentially at different temperatures including first at 100 °C for a time period of 2 hr, followed by 130 °C for a time period of 2 hr, and finally at 150 °C for a time period of 2 hr.

6. DATE AND SIGNATURE:

Dated this 19th day of September, 2024

Patent Agent Name: Hima Bindu Atti

INPA-3925

7. ABSTRACT:

Title: A Method for Synthesis of Bio-Sourced Epoxy Vitrimer for Enhanced Toughness and Recyclability

The present disclosure proposes a method for synthesis of bio-sourced epoxy vitrimer for enhanced toughness and recyclability. The synthesized bio-sourced epoxy vitrimer exhibits enhanced toughness, recyclability, and stress relaxation properties. The synthesized bio-sourced epoxy vitrimer is prepared by partially replacing petrochemical-based components with bio-based alternatives to reduce environmental impact and promote sustainable material solutions. The bio-sourced epoxy vitrimer exhibits ability to be reprocessed and recycled multiple times without significant degradation of material properties. The bio-sourced epoxy vitrimer is configured with enhanced toughness and flexibility compared to traditional epoxies, broadening potential applications. The bio-sourced epoxy vitrimer provides lower carbon footprint and reduced resource depletion.

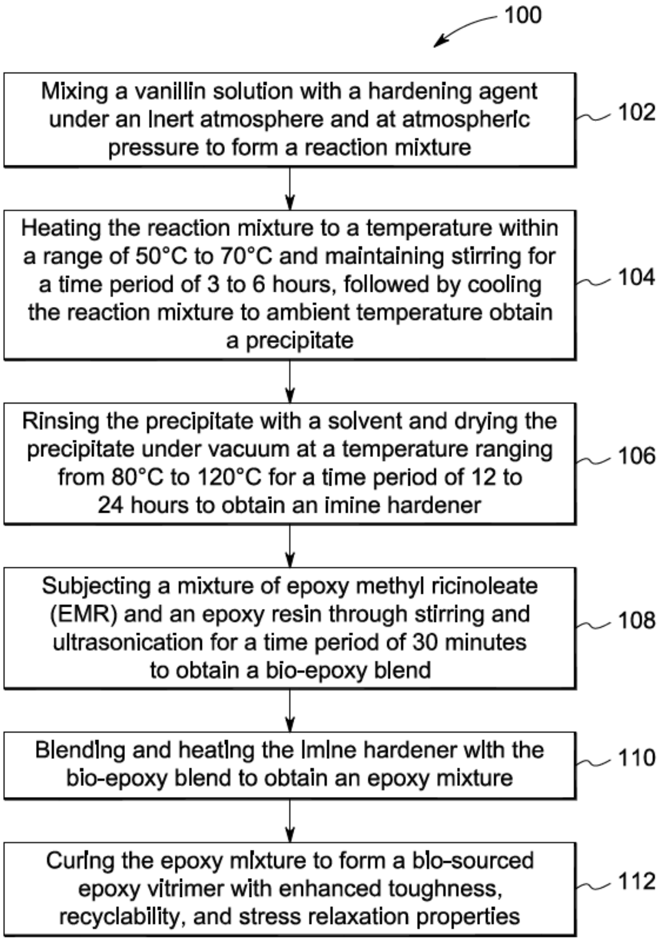


FIG. 1