9, 732-742 (1976). Commercial star block copolymers based on S/I and S/B compositions have been commercialized by companies such as the Phillips Petroleum Company (now part of ConocoPhillips Company, Houston, Texas, United States of America), and BASF (Ludwigshafen, Germany).

Graft copolymers comprising PI backbones and regularly spaced PS side chains attached to the backbone at branch points have also been shown to have elastomeric properties. See, *e.g.*, Uhrig and Mays, *Macromolecules*, 35, 7182-7190 (2002); and Mays et al., *Macromol. Symp.*, 215, 111-126 (2004). However, the synthesis of regular spaced copolymers can be challenging. See Uhrig and Mays, *Macromolecules*, 35, 7182-7190 (2002).

Accordingly, there is need in the art for additional elastomeric polymers and materials that can be produced therefrom, that can be synthesized easily and at low cost. There is also a continuing need for elastomeric polymers that have mechanical properties that can be readily fine-tuned particular end uses.

SUMMARY

In one aspect, the present disclosure refers to a thermoplastic elastomer composition comprising: a random multigraft copolymer comprising: a rubbery polymeric backbone comprising a plurality of randomly spaced branch points, wherein the rubbery polymeric backbone comprises polyisoprene, hydrogenated polyisoprene, polybutadiene, hydrogenated polybutadiene, polyisobutylene, acrylic rubber, or combinations thereof; and a plurality of glassy or semi-crystalline polymeric grafts comprising polystyrene, poly(α-methylstyrene) or another glassy styrenic polymer, polymethylmethacrylate or another glassy acrylic polymer, or combinations thereof, and wherein each of the plurality of glassy or semi-crystalline polymeric grafts is attached to the rubbery polymeric backbone at one of the plurality of randomly spaced branch points; and wherein the random multigraft copolymer has a weight average molecular weight of about 400,000 to 600,00 or more, a volume fraction of glassy or semi-crystalline polymeric graft between about 14% and about 26%, and a strain at break of 1300% or greater.

In another aspect, the present disclosure refers to a fabricated article comprising the thermoplastic elastomer composition as described herein. In yet aspect, the present disclosure refers to an adhesive comprising: a random multigraft

copolymer comprising: a rubbery polymeric backbone comprising a plurality of randomly spaced branch points, wherein the rubbery polymeric backbone comprises polyisoprene, hydrogenated polyisoprene, polybutadiene, hydrogenated polybutadiene, polyisobutylene, acylic rubber, or combinations thereof; and a plurality of glassy or semi-crystalline polymeric grafts comprising polystyrene, poly(α-methylstyrene) or another glassy styrenic polymer, polymethylmethacrylate or another glassy acrylic polymer, or combinations thereof, and wherein each of the plurality of glassy or semi-crystalline polymeric grafts is attached to the rubbery polymeric backbone at one of the plurality of randomly spaced branch points; and wherein the random multigraft copolymer has a weight average molecular weight of about 400,000 to 600,000 or more, a volume fraction of glassy polymeric graft between about 14% and about 26%, and a strain at break of 1300% or greater; and a tackifier.

In yet another aspect, the present disclosure refers to a coated object comprising a coating layer wherein the coating layer comprises the random multigraft copolymer as described herein, wherein the coating layer covers at least a portion of a surface of a wood, ceramic, glass, carbon fiber, metal, metallic, leather, fabric, stone, or plastic object.

In still another aspect, the present disclosure refers to a thermoplastic elastomer composition comprising: a random multigraft copolymer comprising:
a rubbery polymeric backbone comprising a plurality of randomly spaced branch points; and a plurality of glassy or semi-crystalline polymeric grafts, wherein each of the plurality of glassy or semi-crystalline polymeric grafts is attached to the rubbery polymeric backbone at one of the plurality of randomly spaced branch points; and wherein the random multigraft copolymer has a weight average molecular weight of about 400,000 to 600,000 or more, a volume fraction of glassy polymeric graft between about 14% and about 26%, and a strain at break of 1300% or greater.

In some examples, the presently disclosed subject matter provides a thermoplastic elastomer composition comprising a random multigraft copolymer comprising a rubbery polymeric backbone and a plurality of glassy polymeric grafts, wherein each of the plurality of glassy polymeric grafts is attached to the rubbery polymeric backbone at one of a plurality of randomly spaced branch points. In some

CLAIMS

What is claimed is:

1. A thermoplastic elastomer composition comprising:

a random multigraft copolymer comprising:

a rubbery polymeric backbone comprising a plurality of randomly spaced branch points, wherein the rubbery polymeric backbone comprises polyisoprene, hydrogenated polyisoprene, polybutadiene, hydrogenated polybutadiene, polyisobutylene, acrylic rubber, or combinations thereof; and

a plurality of glassy or semi-crystalline polymeric grafts comprising polystyrene, poly(α-methylstyrene) or another glassy styrenic polymer, polymethylmethacrylate or another glassy acrylic polymer, or combinations thereof, and

wherein each of the plurality of glassy or semi-crystalline polymeric grafts is attached to the rubbery polymeric backbone at one of the plurality of randomly spaced branch points; and wherein the random multigraft copolymer has a weight average molecular weight of about 400,000 to 600,000 or more, a volume fraction of glassy or semi-crystalline polymeric graft between about 14% and about 26%, and a strain at break of 1300% or greater.

2. The composition of claim 1, further comprising at least one additional component selected from an organic filler, an inorganic filler, a wax, a tackifier, a plasticizer, an anti-oxidant, a stabilizer, a decorative agent, a biocide, a flame retardant, an anti-static agent, a therapeutic agent or combinations thereof.

3. The composition of claim 2, wherein the decorative agent is a pigment or a dye.

4. The composition of claim 1, wherein the random multigraft copolymer has a residual strain of about 55% or less at an applied strain of about 500% or more.

5. The composition of claim 1, wherein the random multigraft copolymer has a residual strain of about 40% or less at an applied strain of about 300% or more.

6. The composition of claim 1, wherein the random multigraft copolymer has a strain at break of at least 1500%.

7. The composition of claim 1, wherein the random multigraft copolymer has a percent strain at break that increases linearly with the number of branch points.

8. The composition of claim 1, wherein the random multigraft copolymer comprises an architecture selected from a trifunctional comb architecture, a tetrafunctional centipede architecture, or a hexafunctional barbwire architecture.

9. The composition of claim 1, wherein the random multigraft copolymer comprises at least three branch points.

10. A fabricated article comprising the thermoplastic elastomer composition of claim 1.

11. The fabricated article of claim 10, wherein the article is prepared by injection molding, compression molding, extrusion, or calendering.

12. The article of claim 10, wherein the article comprises automotive interior or exterior parts, shoe soles or other shoe parts, elastic waistbands, diaper backings or attachments, adhesive tapes, membranes, balloons, bags, tubing, roofing tiles, medical devices, electronic wiring coatings, or electronic device components.

13. The article of claim 12, wherein the article is a medical device selected from a balloon catheter or a stent.

14. The article of claim 13, wherein the article is a balloon catheter, and wherein at least the inflatable portion of the balloon catheter comprises the thermoplastic elastomer composition.

15. An adhesive comprising:

a random multigraft copolymer comprising:

a rubbery polymeric backbone comprising a plurality of randomly spaced branch points, wherein the rubbery polymeric backbone comprises polyisoprene, hydrogenated polyisoprene, polybutadiene, hydrogenated polybutadiene, polyisobutylene, acylic rubber, or combinations thereof; and

a plurality of glassy or semi-crystalline polymeric grafts comprising polystyrene, poly(α-methylstyrene) or another glassy styrenic polymer, polymethylmethacrylate or another glassy acrylic polymer, or combinations thereof, and

wherein each of the plurality of glassy or semi-crystalline polymeric grafts is attached to the rubbery polymeric backbone at one of the plurality of randomly spaced branch points; and wherein the random multigraft copolymer has a weight average molecular weight of about 400,000 to 600,000 or more, a volume fraction of glassy polymeric graft between about 14% and about 26%, and a strain at break of 1300% or greater; and

a tackifier.

16. The adhesive of claim 15, wherein the random multigraft copolymer comprises an architecture selected from a trifunctional comb architecture, a tetrafunctional centipede architecture, or a hexafunctional barbwire architecture.

17. The adhesive of claim 15, wherein the adhesive is a pressure sensitive adhesive or a hot melt adhesive.

18. The adhesive of claim 15, wherein the tackifier comprises rosins and derivatives thereof, terpenes, modified terpenes, an aliphatic resin, a cycloaliphatic

resin, an aromatic resin, a hydrogenated hydrocarbon resin, a terpene-phenol resin, or mixtures thereof.

19. The adhesive of claim 15, further comprising one or more additives selected from waxes, plasticizers, anti-oxidants, stabilizers, decorative agents, biocides, flame retardants, anti-static agents, or fillers.

20. The adhesive of claim 19, wherein the decorative agent is a pigment, a dye or glitter.

21. A coated object comprising a coating layer wherein the coating layer comprises the random multigraft copolymer of claim 1, wherein the coating layer covers at least a portion of a surface of a wood, ceramic, glass, carbon fiber, metal, metallic, leather, fabric, stone, or plastic object.

22. The coated object of claim 21, wherein the object comprises an article of clothing, an eating/cooking utensil, a medical implant, a medical/surgical tool, or an electronic device.

23. The coated object of claim 21, wherein the object comprises a medical implant or a medical/surgical tool and wherein the coating layer further comprises a therapeutic agent additive selected from the group consisting of a carcinostatic, an immunosuppressive, an antibiotic, an antirheumatic, an antithrombotic, an antihyperlipidemic, an ACE inhibitor, a calcium antagonist, an integrin inhibitor, an antiallergic, an antioxidant, a GPIIb/IIIa antagonist, a retinoid, a flavonoid, a carotenoid, a lipid improving agent, a DNA synthesis inhibitor, a tyrosine kinase inhibitor, an antiplatelet, a vascular smooth muscle antiproliferative agent, an antiinflammatory agent, an interferon, and a NO production accelerator.

24. The coated object of claim 21, wherein the coated object comprises a stent, wherein the stent comprises a metal or metallic wire mesh or a metal or

metallic coil having a cylindrical shape, wherein the wire mesh or coil is covered by the coating layer.

25. A thermoplastic elastomer composition comprising:

a random multigraft copolymer comprising:

a rubbery polymeric backbone comprising a plurality of randomly spaced branch points; and

a plurality of glassy or semi-crystalline polymeric grafts, wherein each of the plurality of glassy or semi-crystalline polymeric grafts is attached to the rubbery polymeric backbone at one of the plurality of randomly spaced branch points; and wherein the random multigraft copolymer has a weight average molecular weight of about 400,000 to 600,000 or more, a volume fraction of glassy polymeric graft between about 14% and about 26%, and a strain at break of 1300% or greater.

26. The thermoplastic elastomer composition of claim 25, wherein the rubbery polymeric backbone has a glass transition temperature of about -40°C or less.

27. The thermoplastic elastomer composition of claim 25, comprising an average of three, four, five, six, seven, eight, or nine branch points.