Claims (Article 34)

1. A biodegradable film, which is prepared from a composition comprising: a copolyester obtained by extrusion reaction of a branched aliphatic-aromatic copolyester derived from monomer a, monomer b, monomer c and monomer d with an organic peroxide, wherein the monomer a is an aromatic dibasic acid or an ester derivative thereof; the monomer b is one or more of C_2-C_{10} aliphatic dihydric alcohols or C_3-C_{10} alicyclic dihydric alcohols; the monomer c is a C_4-C_{20} aliphatic dibasic acid or an ester derivative thereof; the monomer d is one or more of polyols with a functionality of more than 2, polycarboxylic acids with a functionality of more than 2, anhydrides with a functionality of more than 2, epoxy compounds or anhydrides with hydroxyl or carboxyl group, glycidyl ester, glycidyl ether, glycidol, lactones or cyclic carbonates with hydroxyl or carboxyl side group; the branched aliphatic-aromatic copolyester is obtained by reacting monomer a, monomer b, monomer c and monomer d under the action of a catalyst system containing a second catalyst, wherein the second catalyst is a compound of $RE(R^3)_3$, wherein RE is a rare earth metal element, R^3 is at least one selected from the group consisting of halogen, alkoxy group, aryloxy group, acetylacetone group and R^4COO- group, wherein R^4 is C_1-C_{30} alkyl group.

2. The biodegradable film as claimed in claim 1, wherein, in the compound of $RE(R^3)_3$ as the second catalyst, RE is lanthanum, cerium, praseodymium, neodymium, terbium, ytterbium, dysprosium, samarium or scandium; the halogen is chlorine or bromine, the alkoxy group is C_3-C_6 alkoxy group, the aryloxy group is aryloxy group comprising at least one benzene ring and/or naphthalene ring, and R^4 is C_1-C_{20} alkyl group.

3. The biodegradable film as claimed in claim 1, wherein the catalyst system further comprises a first catalyst and/or a third catalyst; wherein the first

catalyst is one or more selected from the group consisting of an oxide of M, $M(OR^1)_n$ and $M(OOCR^2)_m$, wherein M is titanium, antimony or zinc, n and m are each independently the valence of M, R^1 is C_1-C_{10} alkyl group, and R^2 is C_1-C_{20} alkyl group; the third catalyst is at least one organotin compound.

4. The biodegradable film as claimed in claim 3, wherein the molar ratio of the catalyst in its entirety to the monomer (a + c) is 1: 1000-20000, and the molar ratio of the first catalyst to the second catalyst to the third catalyst is 0-20: 0.1-10: 0-1, preferably 0.1-20: 0.1-10: 1.

5. The biodegradable film as claimed in any one of claims 1-4, wherein the amounts of various monomers are as follows: the molar ratio of (a + c) to b is 1: 0.8-3, the molar ratio of (a + c) to d is 100-2000: 1, the molar ratio of a to c is 0: 100-95: 5, preferably 0: 100-60: 40.

6. The biodegradable film as claimed in any one of claims 1-5, wherein the monomer a is selected from terephthalic acid or dimethyl terephthalate; the monomer b is selected from ethylene glycol, 1,3-propanediol or 1,4-butanediol; the monomer c is selected from succinic acid, succinic anhydride, dimethyl succinate, adipic acid or dimethyl adipate; the monomer d is selected from pyromellitic dianhydride, glycerol, pentaerythritol, glycidyl succinate, glycidyl adipate, glycidyl terephthalate, ethylene glycol glycidyl ether, 1,4-butanediol glycidyl ether, glycidic acid, glycidol, 3-hydroxyphthalic anhydride, 4-hydroxyphthalic anhydride, 3-hydroxymethylcaprolactone, a five-membered or six-membered cyclic carbonate with an alkylhydroxy group or an alkylcarboxyl group.

7. The biodegradable film as claimed in any one of claims 1-6, wherein the branched aliphatic-aromatic copolyester is prepared as follows: under the action

of a catalyst system, monomer a, monomer b, monomer c and monomer d are mixed for esterification or transesterification reaction, or the esterification or transesterification product of monomer a and monomer b and the esterification or transesterification product of monomer c and monomer d are mixed for copolycondensation reaction.

8. The biodegradable film as claimed in any one of claims 1-7, wherein the branched aliphatic-aromatic copolyester has a melt index of 2-100g/10min, preferably 5-100g/10min at 190℃ under a load of 2.16 kg.

9. The biodegradable film as claimed in any one of claims 1-8, wherein the organic peroxide is selected from dialkyl peroxides, diacyl peroxides or peroxyesters.

10. The biodegradable film as claimed in claim 9, wherein the dialkyl peroxide is selected from dicumyl peroxide, 2,5-dimethyl-2,5-di(tert-butylperoxy)hexane or di(tert-butylperoxyisopropyl)benzene; the diacyl peroxide is selected from benzoyl peroxide or lauroyl peroxide.

11. The biodegradable film as claimed in any one of claims 1-10, wherein the organic peroxide is used in an amount of 0.01 to 5wt% based on the amount of the branched aliphatic-aromatic copolyester.

12. The biodegradable film as claimed in any one of claims 1-11, wherein the copolyester has a melt index of 0.1 to 10g/10min at 190℃ under a load of 2.16 kg.

13. The biodegradable film as claimed in any one of claims 1-12, wherein

the composition also comprises additives, and the additives include an antioxidant, a slipping agent, an anti-blocking agent and a nucleating agent.

14. The biodegradable film as claimed in claim 13, wherein the amount of the antioxidant is 0.05-5 parts, the amount of the slipping agent is 0.05-5 parts, the amount of the anti-blocking agent is 0.05-5 parts, and the amount of the nucleating agent is 0.1-20 parts based on 100 parts by weight of the copolyester.

15. The biodegradable film as claimed in any one of claims 13-14, wherein the antioxidant is a mixture of a hindered phenol antioxidant and a phosphite antioxidant in a mass ratio of 1: 1.

16. The biodegradable film as claimed in claim 15, wherein the hindered phenol antioxidant is selected from antioxidant 1010, antioxidant 1076, antioxidant 245 or antioxidant 246; the phosphite antioxidant is selected from triphenyl phosphate, trimethyl phosphate or antioxidant 168.

17. The biodegradable film as claimed in any one of claims 13-14, wherein the slipping agent is stearate or organic carboxylic acid amide, wherein the stearate is preferably calcium stearate, and the organic carboxylic acid amide is preferably erucamide or oleamide;

the anti-blocking agent is silica or talc;

the nucleating agent is selected from hyperbranched polyamides, low density polyethylene or ethylene-methacrylic acid ionomers.

18. A method for preparing the biodegradable film as claimed in any one of claims 1-17, the method comprising the steps of:

1) preparing a copolyester by extrusion reaction of a branched aliphatic-aromatic copolyester derived from monomer a, monomer b, monomer

c and monomer d with an organic peroxide;

the monomer a is an aromatic dibasic acid or an ester derivative thereof; the monomer b is one or more of C_2-C_{10} aliphatic dihydric alcohols or C_3-C_{10} alicyclic dihydric alcohols; the monomer c is a C_4-C_{20} aliphatic dibasic acid or an ester derivative thereof; the monomer d is one or more of polyols with a functionality of more than 2, polycarboxylic acids with a functionality of more than 2, anhydrides with a functionality of more than 2, epoxy compounds or anhydrides with hydroxyl or carboxyl group, glycidyl ester, glycidyl ether, glycidol, lactones or cyclic carbonates with hydroxyl or carboxyl side group;

the branched aliphatic-aromatic copolyester is obtained by reacting monomer a, monomer b, monomer c and monomer d under the action of a catalyst system containing a second catalyst, wherein the second catalyst is a compound of $RE(R^3)_3$, wherein RE is a rare earth metal element, R^3 is at least one selected from the group consisting of halogen, alkoxy group, aryloxy group, acetylacetone group and R^4COO- group, wherein R^4 is C_1-C_{30} alkyl group;

2) extruding and granulating the copolyester obtained in the step 1) to obtain a composition; and

3) blowing the composition to obtain the biodegradable film.

19. The method as claimed in claim 18, wherein, in the compound of $RE(R^3)_3$ as the second catalyst, RE is lanthanum, cerium, praseodymium, neodymium, terbium, ytterbium, dysprosium, samarium or scandium; the halogen is chlorine or bromine, the alkoxy group is C_3-C_6 alkoxy group, the aryloxy group is aryloxy group comprising at least one benzene ring and/or naphthalene ring, and R^4 is C_1-C_{20} alkyl group.

20. The method as claimed in claim 18, wherein the catalyst system further comprises a first catalyst and/or a third catalyst; wherein the first catalyst is one or more selected from the group consisting of an oxide of M, $M(OR^1)_n$ and

$M(OOCR^2)_m$, wherein M is titanium, antimony or zinc, n and m are each independently the valence of M, R^1 is C_1-C_{10} alkyl group, and R^2 is C_1-C_{20} alkyl group; the third catalyst is at least one organotin compound.

21. The method as claimed in claim 20, wherein the molar ratio of the catalyst in its entirety to the monomer (a + c) is 1: 1000-20000, and the molar ratio of the first catalyst to the second catalyst to the third catalyst is 0-20: 0.1-10: 0-1, preferably 0.1-20: 0.1-10: 1.

22. The method as claimed in any one of claims 18-21, wherein the amounts of various monomers are as follows: the molar ratio of (a + c) to b is 1: 0.8-3, the molar ratio of (a + c) to d is 100-2000: 1, the molar ratio of a to c is 0: 100-95: 5, preferably 0: 100-60: 40.

23. The method as claimed in any one of claims 18-22, wherein the monomer a is selected from terephthalic acid or dimethyl terephthalate; the monomer b is selected from 1,3-propanediol or 1,4-butanediol; the monomer c is selected from succinic acid, succinic anhydride, dimethyl succinate, adipic acid or dimethyl adipate; the monomer d is selected from pyromellitic dianhydride, glycerol, pentaerythritol, glycidyl succinate, glycidyl adipate, glycidyl terephthalate, ethylene glycol glycidyl ether, 1,4-butanediol glycidyl ether, glycidic acid, glycidol, 3-hydroxyphthalic anhydride, 4-hydroxyphthalic anhydride, 3-hydroxymethylcaprolactone, a five-membered or six-membered cyclic carbonate with an alkylhydroxy group or an alkylcarboxyl group.

24. The method as claimed in any one of claims 18-23, wherein the branched aliphatic-aromatic copolyester is prepared as follows: under the action of a catalyst system, monomer a, monomer b, monomer c and monomer d are mixed for esterification or transesterification reaction, or the esterification or

transesterification product of monomer a and monomer b and the esterification or transesterification product of monomer c and monomer d are mixed for copolycondensation reaction.

25. The method as claimed in any one of claims 18-24, wherein the branched aliphatic-aromatic copolyester has a melt index of 2-100g/10min, preferably 5-100g/10min at 190℃ under a load of 2.16 kg.

26. The method as claimed in any one of claims 18-25, wherein the extrusion temperature in the step 1) is 130-190℃, preferably 150-180℃, and more preferably 160-180℃.

27. The method as claimed in any one of claims 18-26, wherein the organic peroxide is selected from dialkyl peroxides, diacyl peroxides or peroxyesters.

28. The method as claimed in claim 27, wherein the dialkyl peroxide is selected from dicumyl peroxide, 2,5-dimethyl-2,5-di(tert-butylperoxy)hexane or di(tert-butylperoxyisopropyl)benzene; the diacyl peroxide is selected from benzoyl peroxide or lauroyl peroxide.

29. The method as claimed in any one of claims 18-28, wherein the organic peroxide is used in an amount of 0.01 to 5wt% based on the amount of the branched aliphatic-aromatic copolyester.

30. The method as claimed in any one of claims 18-29, wherein the copolyester has a melt index of 0.1 to 10g/10min at 190℃ under a load of 2.16 kg.

31. The method as claimed in any one of claims 18-30, wherein the copolyester is also mixed with additives in the step 2), wherein the additives include an antioxidant, a slipping agent, an anti-blocking agent and a nucleating agent.

32. The method as claimed in claim 31, wherein the amount of the antioxidant is 0.05-5 parts, the amount of the slipping agent is 0.05-5 parts, the amount of the anti-blocking agent is 0.05-5 parts, and the amount of the nucleating agent is 0.1-20 parts based on 100 parts by weight of the copolyester.

33. The method as claimed in any one of claims 31-32, wherein the antioxidant is a mixture of a hindered phenol antioxidant and a phosphite antioxidant in a mass ratio of 1: 1.

34. The method as claimed in claim 33, wherein the hindered phenol antioxidant is selected from antioxidant 1010, antioxidant 1076, antioxidant 245 or antioxidant 246; the phosphite antioxidant is selected from triphenyl phosphate, trimethyl phosphate or antioxidant 168.

35. The method as claimed in any one of claims 31-32, wherein the slipping agent is stearate or organic carboxylic acid amide, wherein the stearate is preferably calcium stearate, and the organic carboxylic acid amide is preferably erucamide or oleamide;

the anti-blocking agent is silica or talc;

the nucleating agent is selected from hyperbranched polyamides, low density polyethylene or ethylene-methacrylic acid ionomers.

36. A copolyester, which is obtained by extrusion reaction of a branched aliphatic-aromatic copolyester derived from monomer a, monomer b, monomer

c and monomer d with an organic peroxide, wherein the monomer a is an aromatic dibasic acid or an ester derivative thereof; the monomer b is one or more of C_2-C_{10} aliphatic dihydric alcohols or C_3-C_{10} alicyclic dihydric alcohols; the monomer c is a C_4-C_{20} aliphatic dibasic acid or an ester derivative thereof; the monomer d is one or more of polyols with a functionality of more than 2, polycarboxylic acids with a functionality of more than 2, anhydrides with a functionality of more than 2, epoxy compounds or anhydrides with hydroxyl or carboxyl group, glycidyl ester, glycidyl ether, glycidol, lactones or cyclic carbonates with hydroxyl or carboxyl side group; the branched aliphatic-aromatic copolyester is obtained by reacting monomer a, monomer b, monomer c and monomer d under the action of a catalyst system containing a second catalyst, wherein the second catalyst is a compound of $RE(R^3)_3$, wherein RE is a rare earth metal element, R^3 is at least one selected from the group consisting of halogen, alkoxy group, aryloxy group, acetylacetone group and R^4COO- group, wherein R^4 is C_1-C_{30} alkyl group.

37. The copolyester as claimed in claim 36, wherein, in the compound of $RE(R^3)_3$ as the second catalyst, RE is lanthanum, cerium, praseodymium, neodymium, terbium, ytterbium, dysprosium, samarium or scandium; the halogen is chlorine or bromine, the alkoxy group is C_3-C_6 alkoxy group, the aryloxy group is aryloxy group comprising at least one benzene ring and/or naphthalene ring, and R^4 is C_1-C_{20} alkyl group.

38. The copolyester as claimed in claim 36, wherein the catalyst system further comprises a first catalyst and/or a third catalyst; wherein the first catalyst is one or more selected from the group consisting of an oxide of M, $M(OR^1)_n$ and $M(OOCR^2)_m$, wherein M is titanium, antimony or zinc, n and m are each independently the valence of M, R^1 is C_1-C_{10} alkyl group, and R^2 is C_1-C_{20} alkyl group; the third catalyst is at least one organotin compound.

39. The copolyester as claimed in claim 38, wherein the molar ratio of the catalyst in its entirety to the monomer (a + c) is 1: 1000-20000, and the molar ratio of the first catalyst to the second catalyst to the third catalyst is 0-20: 0.1-10: 0-1, preferably 0.1-20: 0.1-10: 1.

40. The copolyester as claimed in any one of claims 36-39, wherein the amounts of various monomers are as follows: the molar ratio of (a + c) to b is 1: 0.8-3, the molar ratio of (a + c) to d is 100-2000: 1, the molar ratio of a to c is 0: 100-95: 5, preferably 0: 100-60: 40.

41. The copolyester as claimed in any one of claims 36-40, wherein the monomer a is selected from terephthalic acid or dimethyl terephthalate; the monomer b is selected from ethylene glycol, 1,3-propanediol or 1,4-butanediol; the monomer c is selected from succinic acid, succinic anhydride, dimethyl succinate, adipic acid or dimethyl adipate; the monomer d is selected from pyromellitic dianhydride, glycerol, pentaerythritol, glycidyl succinate, glycidyl adipate, glycidyl terephthalate, ethylene glycol glycidyl ether, 1,4-butanediol glycidyl ether, glycidic acid, glycidol, 3-hydroxyphthalic anhydride, 4-hydroxyphthalic anhydride, 3-hydroxymethylcaprolactone, a five-membered or six-membered cyclic carbonate with an alkylhydroxy group or an alkylcarboxyl group.

42. The copolyester as claimed in any one of claims 36-41, wherein the branched aliphatic-aromatic copolyester is prepared as follows: under the action of a catalyst system, monomer a, monomer b, monomer c and monomer d are mixed for esterification or transesterification reaction, or the esterification or transesterification product of monomer a and monomer b and the esterification or transesterification product of monomer c and monomer d are mixed for

copolycondensation reaction.

43. The copolyester as claimed in any one of claims 36-42, wherein the branched aliphatic-aromatic copolyester has a melt index of 2-100g/10min, preferably 5-100g/10min at 190℃ under a load of 2.16 kg.

44. The copolyester as claimed in any one of claims 36-43, wherein the organic peroxide is selected from dialkyl peroxides, diacyl peroxides or peroxyesters.

45. The copolyester as claimed in claim 44, wherein the dialkyl peroxide is selected from dicumyl peroxide, 2,5-dimethyl-2,5-di(tert-butylperoxy)hexane or di(tert-butylperoxyisopropyl)benzene; the diacyl peroxide is selected from benzoyl peroxide or lauroyl peroxide.

46. The copolyester as claimed in any one of claims 36-45, wherein the organic peroxide is used in an amount of 0.01 to 5wt% based on the amount of the branched aliphatic-aromatic copolyester.

47. The copolyester as claimed in any one of claims 36-46, wherein the copolyester has a melt index of 0.1 to 10g/10min at 190℃ under a load of 2.16 kg.