CLAIMS:

1. A method for chlorinating a carbohydrate or a derivative thereof to produce a polychlorinated carbohydrate or a derivative thereof, said polychlorinated carbohydrate or a derivative thereof having a desired number of chlorine atoms in desired locations in its molecular structure, the method comprising:

(i) reacting the carbohydrate or derivative thereof with a chlorinating agent to obtain a reaction mixture comprising said polychlorinated carbohydrate or derivative thereof and at least one under-chlorinated carbohydrate or derivative thereof;

(ii) returning the at least one under-chlorinated carbohydrate or derivative thereof to a chlorinating step and further chlorinating the at least one under-chlorinated carbohydrate or derivative thereof to obtain the desired polychlorinated carbohydrate or derivative thereof; and

(iii) optionally repeating steps (i) and (ii) "n" times where $n \geq 1$.

2. The method of claim 1, further comprising:

(iv) quenching at least a portion of the reaction mixture from step (i) to obtain a quenched reaction mixture and

(v) isolating a product mixture comprising polychlorinated carbohydrate or derivative thereof and at least one under-chlorinated carbohydrate or derivative thereof from the quenched reaction mixture.

3. The method of claim 1, wherein the at least one under-chlorinated carbohydrate or derivative thereof is returned to step (i) for further chlorination.

4. The method of claim 1, wherein the further chlorination of the at least one under-chlorinated carbohydrate or derivative thereof takes place in a vessel separate from the vessel where step (i) takes place.

5. The method of claim 1, wherein the further chlorination of the at least one under-chlorinated carbohydrate or derivative thereof takes place in the same vessel where step (i) takes place.

6. The method of any one of claims 1 to 5, which is a continuous feed method.

7. The method of claim 2, wherein in step (v), the product mixture is isolated from the quenched reaction mixture by solvent extraction.

8. The method of claim 2 or 7, further comprising: (vi) separating the product mixture isolated in step (v) into (a) a first fraction comprising the polychlorinated carbohydrate or derivative thereof and (b) a second fraction comprising at least one under-chlorinated carbohydrate or derivative thereof.

9. The method of any one of claims 1 to 8, wherein the polychlorinated carbohydrate or derivative thereof, under-chlorinated carbohydrate or derivative thereof, or a mixture thereof, is optionally combined with additional carbohydrate or derivative thereof and returned to a chlorinating step or to step (i).

10. The method of any one of claims 1 to 9, wherein the carbohydrate or derivative thereof is a sugar or derivative thereof.

11. The method of claim 10, wherein the sugar derivative is a sugar ester.

12. The method of claim 11, wherein the sugar ester is a sucrose-6-ester.

13. The method of any one of claims 1 to 12, wherein the polychlorinated carbohydrate derivative is 4,1',6'-trichloro-4,1',6'-trideoxy-6-*O*-ester of galactosucrose (TGS-6E).

14. The method of any one of claims 11 to 13, wherein the polychlorinated carbohydrate ester or the under-chlorinated ester is of the formula:

M-OC(=O) R',

wherein R' comprises a hydrophobic group or a hydrophobic polymer and M is the moiety completing the rest of the polychlorinated carbohydrate ester or the under-chlorinated ester.

15. The method of claim 14, wherein R' comprises a hydrophobic group or a hydrophobic polymer such that the polychlorinated carbohydrate ester or the under-chlorinated ester has an octanol-water partition coefficient cLogP of -0.5 or greater.

16. The method of claim 14, wherein the polychlorinated carbohydrate ester or the under-chlorinated carbohydrate ester has an octanol-water partition coefficient cLogP of 0 or greater than 0.

17. The method of claim 14, wherein R is alkyl, aryl, or arylalkyl, wherein the aryl part of aryl or arylalkyl is optionally substituted with one or more substituents selected from the group consisting of halo, haloalkyl, cyano, nitro, alkoxy, alkylthio, acyl, acyloxy, thioacyl, acylthio, and aryloxy.

18. The method of claim 14, wherein R' is aryl.

19. The method of claim 18, wherein R' is phenyl.

20. The method of claim 14, wherein R' comprises a hydrophobic polymer.

21. The method of any one of claims 1 to 20, wherein the chlorinating agent is an acid chloride or an activated acid chloride.

22. The method of claim 21, wherein the acid chloride is selected from the group consisting of thionyl chloride, sulfuryl chloride, phosgene, phosphorus pentachloride, oxalyl chloride, methane sulfonyl chloride, and bis(trichloromethyl)carbonate.

23. The method of claim 21, wherein the activated acid chloride is a Vilsmeier Reagent [or Arnold's reagent] having the formula: $[XYC=N^{+}R_2]\ Cl^{-}$, wherein X is hydrogen, aryl, or alkyl, wherein the aryl or alkyl is optionally substituted with a halogen, alkoxy, thioalkoxy, amido, or cyano; Y is a leaving group; and R is hydrogen or alkyl which is optionally substituted with halogen, alkoxy, thioalkoxy, amido, or cyano.

24. The method of claim 23, wherein Y is halogen, heteroalkyl, or a group capable of being displaced by a heteroatomic nucleophile.

25. The method of claim 24, wherein Y is halogen.

26. The method of claim 23, wherein X is hydrogen.

27. The method of claim 23, wherein R and Y are alkyl.

28. The method of any one of claims 23 to 26, wherein X is hydrogen, Y is chloro, and R is methyl.

29. The method of claim 28, wherein the Vilsmeier Reagent is produced by the reaction of N,N-dimethylformamide with an acid chloride.

30. The method of claim 29, wherein the acid chloride is selected from the group consisting of thionyl chloride, sulfuryl chloride, phosgene, phosphorus pentachloride, oxalyl chloride, methane sulfonyl chloride, and bis(trichloromethyl)carbonate.

31. The method of any one of claims 1 to 30, wherein the chlorination reaction is carried out in a solvent system.

32. The method of claim 31, wherein the solvent system comprises one or more polar solvents, one or more non-polar solvents, or a mixture thereof.

33. The method of claim 32, wherein the polar solvent is a polar aprotic solvent.

34. The method of claim 33, wherein the polar aprotic solvent is selected from the group consisting of N,N-dimethylformamide, N,N-dimethylacetamide, N-methyl pyrrolidone, dimethylsulfoxide, sulfolane, and glycol ethers.

35. The method of claim 31, wherein the solvent system comprises a mixture of a polar aprotic solvent and at least one other aprotic solvent selected from the group consisting of chlorinated hydrocarbons and ethers.

36. The method of claim 35, wherein the chlorinated hydrocarbon is selected from the group consisting of chloroform, dichloromethane, dichloroethane, chlorofluorocarbons, dichloroethylene, trichloroethylene, trichloropropane, trichloroethane, dichloroethane, tetrachloroethane, and perfluorooctane.

37. The method of claim 35, wherein the ether is selected from the group consisting of tetrahydrofuran, dioxane, methoxyethane, dimethoxymethane, dimethoxyethane, tetrahydropyran, diglyme, diisopropyl ether, diethyl ether, and methyl t-butyl ether.

38. The method of any one of claims 32 to 37, wherein the solvent system comprises N,N-dimethylformamide as the polar aprotic solvent.

39. A method for chlorinating a carbohydrate or a derivative thereof to obtain a polychlorinated carbohydrate or derivative thereof, said polychlorinated carbohydrate or derivative thereof having a desired number of chlorine atoms in desired locations in its molecular structure, the method comprising:

(i) dissolving the carbohydrate or derivative thereof in an aprotic solvent system to obtain a solution comprising the carbohydrate or derivative thereof;

(ii) combining the solution comprising the carbohydrate or derivative thereof from step (i) with a chlorinating agent to obtain a chlorination mixture;

(iii) heating the chlorination mixture to obtain a mixture of chlorinated carbohydrates or derivatives thereof comprising the desired polychlorinated carbohydrate or derivative thereof and at least one under-chlorinated carbohydrate or derivative thereof having less than the desired number of chlorine atoms;

(iv) returning at least one under-chlorinated carbohydrate or derivative thereof to a chlorinating step so as to further chlorinate the at least one under-chlorinated carbohydrate or derivative thereof; and optionally

(v) repeating steps (ii)-(iv) "n" times where $n \geq 1$.

40. The method of claim 39, further comprising (vi) separating the polychlorinated carbohydrate or derivative thereof from the at least one under-chlorinated carbohydrates or derivatives thereof.

41. The method of claim 39, wherein the at least one under-chlorinated carbohydrate or derivative thereof is returned to step (i) for further chlorination.

42. The method of claim 39, wherein the further chlorination of the at least one under-chlorinated carbohydrate or derivative thereof takes place in a vessel separate from the vessel where step (i) takes place.

43. The method of claim 39, wherein the further chlorination of the at least one under-chlorinated carbohydrate or derivative thereof takes place in the same vessel where step (i) takes place.

44. The method of any one of claims 39 to 43, which is a continuous feed method.

45. The method of any one of claims 1 to 44, wherein the polychlorinated carbohydrate or derivative thereof is a polychlorinated sucrose-6-ester.

46. The method of claim 45, wherein the polychlorinated carbohydrate is sucralose-6-ester.

47. The method of claim 46, wherein the sucralose-6-ester is obtained in step (i) in an yield of at least 30%.

48. The method of claim 47, wherein the sucralose-6-ester is obtained in step (i) in an yield of at least 40%.

49. The method of claim 48, wherein the sucralose-6-ester is obtained in step (i) in an yield of at least 50%.

50. The method of claim 39, wherein the sucralose-6-ester is obtained in step (v) in an yield of at least 50%.

51. The method of claim 50, wherein sucralose-6-ester is obtained in step (v) an yield of the yield of at least 60%.

52. The method of claim 51, wherein the sucralose-6-ester is obtained in step (v) in an yield of at least 70%.

53. The method of claim 52, wherein sucralose-6-ester is obtained in step (v) an yield of the yield of at least 80%.

54. The method of claim 53, wherein sucralose-6-ester is obtained in step (v) an yield of the yield of at least 90%.

55. The method of claim 1, wherein the polychlorinated carbohydrate or derivative thereof is obtained in step (i) in an yield of at least 30%.

56. The method of claim 55, wherein the polychlorinated carbohydrate or derivative thereof is obtained in step (i) in an yield of at least 40%.

57. The method of claim 56, wherein the polychlorinated carbohydrate or derivative thereof is obtained in step (i) in an yield of at least 50%.

58. The method of claim 1, wherein the polychlorinated carbohydrate or derivative thereof is obtained in step (iii) in an yield of at least 50%.

59. The method of claim 58, wherein polychlorinated carbohydrate or derivative thereof is obtained in step (iii) in an yield of the yield of at least 60%.

60. The method of claim 59, wherein the polychlorinated carbohydrate or derivative thereof is obtained in step (iii) in an yield of at least 70%.

61. The method of claim 60, wherein polychlorinated carbohydrate or derivative thereof is obtained in step (iii) in an yield of the yield of at least 80%.

62. The method of claim 61, wherein polychlorinated carbohydrate or derivative thereof is obtained in step (iii) in an yield of the yield of at least 90%.

63. A method of preparing sucralose comprising de-esterifying the polychlorinated sucrose 6-ester obtained according to any one of claims 1 to 62 to obtain sucralose.

64. The method of any one of claims 1 to 62, wherein the under-chlorinated carbohydrate or derivative thereof has one or two chlorine atoms in its molecular structure.

65. The method of claim 1 or 39, wherein the desired number of chlorine atoms present in the polychlorinated carbohydrate or derivative thereof is 3.

CLAIMS

1. A method for chlorinating a sucrose-6-ester to produce 4,1',6'-trichloro-4,1',6'-trideoxy-6-*O*-ester of galactosucrose (TGS-6E) of the formula M-OC(=O) R', wherein R' is a hydrophobic group such that TGS-6E has an octanol-water partition coefficient cLogP of 0.5 or greater, and M is the moiety completing the rest of TGS-6E, the method comprising:

(i) reacting the sucrose-6-ester with a chlorinating agent in N,N-dimethylformamide (DMF) to obtain a chlorination reaction mixture, and removing at least a portion of the HCl resulting from the chlorination reaction as a DMF-HCl mixture by vacuum distillation, to obtain a reaction mixture comprising said TGS-6E and at least one under-chlorinated sucrose-6-ester of the formula M'-OC(=O) R', wherein R' is a hydrophobic group such that the under-chlorinated sucrose-6-ester has an octanol-water partition coefficient cLogP of 0.5 or greater, and M' is the moiety completing the rest of the under-chlorinated sucrose-6- ester;

(ii) returning the at least one under-chlorinated sucrose-6-ester to a chlorinating step and further chlorinating the at least one sucrose-6-ester to obtain TGS-6E; and

(iii) optionally repeating steps (i) and (ii) "n" times where n $\geq$1.

2. The method of claim 1, further comprising:

(iv) quenching at least a portion of the reaction mixture from step (i) to obtain a quenched reaction mixture and

(v) isolating a product mixture comprising TGS-6E and at least one under-chlorinated sucrose-6-ester from the quenched reaction mixture.

3. The method of claim 1, wherein;

(a) the at least one under-chlorinated sucrose-6-ester is returned to step (i) for further chlorination; or

(b) wherein the further chlorination of the at least one under-chlorinated sucrose-6-ester takes place in a vessel separate from the vessel where step (i) takes place; or

(c) wherein the further chlorination of the at least one under-chlorinated sucrose-6-ester takes place in the same vessel where step (i) takes place.

4 The method of claim 2, wherein in step (v), the product mixture is isolated from the quenched reaction mixture by solvent extraction; and/or the method comprises (vi) separating the product mixture isolated in step (v) into (a) a first fraction comprising TGS-6E and (b) a second fraction comprising at least one under-chlorinated sucrose-6-ester.

5. The method of any one of claims 1 to 4, wherein TGS-6E, the at least one under-chlorinated sucrose-6-ester, or a mixture thereof, is optionally combined with additional sucrose-6-ester and returned to a chlorinating step or to step (i).

6. The method of any one of claims 1 to 5, wherein R' is phenyl.

7. The method of any one of claims 1 to 6, wherein the chlorinating agent is an acid chloride, an activated acid chloride, or bis(trichloromethyl)carbonate, preferably wherein the acid chloride is selected from the group consisting of thionyl chloride, sulfuryl chloride, phosgene, phosphorus pentachloride, oxalyl chloride, and methanesulfonyl chloride, and wherein the activated acid chloride is a Vilsmeier Reagent having the formula: $[XYC=N^{+}R_2]$ Cl^{-}, wherein X is hydrogen, aryl, or alkyl, wherein the aryl or alkyl is optionally substituted with a halogen, alkoxy, thioalkoxy, amido, or cyano; Y is a leaving group, preferably Y is halogen, heteroalkyl, or a group capable of being displaced by a heteroatomic nucleophile; and R is hydrogen or alkyl which is optionally substituted with halogen, alkoxy, thioalkoxy, amido, or cyano.

8. The method of claim 7, wherein X is hydrogen, Y is chloro, and R is methyl.

9. The method according to claim 1, wherein the TGS-6E is sucrose-6-benzoate ester (TGS-6B), the method comprising:

(a) dissolving the sucrose-6-benzoate ester in N,N-dimethylformamide (DMF) to obtain a solution comprising the sucrose-6-benzoate ester;

(b) combining the solution comprising the sucrose-6-benzoate ester from step (i) with a chlorinating agent to obtain a chlorination mixture;

(c) removing by vacuum distillation at least a portion of the HCl resulting from the chlorination reaction as a DMF-HCl mixture to obtain a reaction mixture comprising TGS-6B and at least one under-chlorinated sucrose-6-benzoate ester;

(d) returning the at least one under-chlorinated sucrose-6-benzoate ester to a chlorinating step so as to further chlorinate the at least one under-chlorinated sucrose-6-benzoate ester; and optionally

(e) repeating steps (b)-(d) "n" times where n $\geq$1.

10. The method of claim 9, further comprising (f) separating the TGS-6B from the at least one under-chlorinated sucrose-6-benzoate ester.

11. The method of claim 9, wherein the at least one under-chlorinated sucrose-6-benzoate ester is returned to step (a) for further chlorination.

12. The method of claims 1 to 11, wherein n = 2 to 8.

13. The method of claim 1 or 9, wherein the at least one under-chlorinated sucrose-6-ester is dichlorogalactosucrose-6-benzoate.

14. The method of claim 1 or 9, wherein the at least one under-chlorinated sucrose-6-ester is monochlorosucrose-6-benzoate.