WHAT IS CLAIMED IS:

1. A pretreatment composition for treating a metal substrate comprising:

a Group IIIB and/or Group IVB metal present in an amount of 50 ppm to 500 ppm based on a total weight of the ingredients in the pretreatment composition;

free fluoride;

lithium; and

an electropositive metal;

wherein a molar ratio of the Group IIIB and/or Group IV metal to the lithium is 100:1 to 1:10; and

wherein the composition is substantially free of phosphate ions.

2. The pretreatment composition of claim 1, wherein the pretreatment composition comprises a Group IVB metal.

3. The pretreatment composition of claim 2, wherein the Group IVB metal is provided in the form of hexafluorozirconic acid, hexafluorotitanic acid, or salts thereof.

4. The pretreatment composition of claim 2, wherein the Group IVB metal is zirconium.

5. The pretreatment composition according to claim 1, wherein the Group IVB metal is provided in the form of oxides or hydroxides of zirconium.

6. The pretreatment composition according to claim 1, wherein the Group IVB metal is provided in the form of zirconyl nitrate, zirconyl sulfate, or zirconium basic carbonate.

7. The pretreatment composition of claim 1, wherein the Group IIIB and/or Group IVB metal is provided in the form of an acid or salt.

8. The pretreatment composition of claim 1, wherein the pretreatment composition is substantially free of resinous binder.

18. The pretreatment composition of claim 1, wherein the pretreatment composition is aqueous.

19. The pretreatment composition of claim 1, wherein the pretreatment composition is used in a dip application.

20. The pretreatment composition of claim 1, wherein the pretreatment composition is used in a spray application.

21. The pretreatment composition of claim 1, wherein a K ratio is equal to A/B, wherein A is a mole weight of a compound (A) containing the Group IIIB and/or Group IVB metal, and wherein B is a mole weight calculated as HF of a compound containing fluorine as a supplying source of the fluoride, wherein $K>0.10$.

22. The pretreatment composition of claim 1, wherein a K ratio is equal to A/B, wherein A is a mole weight of a compound (A) containing the Group IIIB and/or Group IVB metal, and wherein B is a mole weight calculated as HF of a compound containing fluorine as a supplying source of the fluoride, wherein $0.11<K<0.25$.

23. The pretreatment composition of claim 1, wherein the electropositive metal is selected from the Group consisting of copper, nickel, silver, gold, and combinations thereof.

24. The pretreatment composition of claim 1, wherein the electropositive metal comprises copper.

25. The pretreatment composition of claim 24, wherein the copper is provided in the form of copper nitrate, copper sulfate, copper chloride, copper carbonate, or copper fluoride.

26. The pretreatment composition of claim 1, wherein the electropositive metal comprises from 0 to 100 parts per million, based on a total weight of the ingredients in the pretreatment composition.

27. The pretreatment composition of claim 1, wherein the electropositive metal comprises from 2 to 35 parts per million, based on a total weight of the ingredients in the pretreatment composition.

28. The pretreatment composition of claim 1, further comprising molybdenum.

29. The pretreatment composition of claim 28, wherein the molybdenum is provided in the form of a salt.

30. The pretreatment composition of claim 29, wherein the salt is sodium molybdate, calcium molybdate, potassium molybdate, ammonium molybdate, molybdenum chloride, molybdenum acetate, molybdenum sulfamate, molybdenum formate, or molybdenum lactate.

31. The pretreatment composition of claim 28, wherein the molybdenum comprises from 5 to 500 parts per million, based on a total weight of the ingredients in the pretreatment composition.

32. The pretreatment composition of claim 28, wherein the molybdenum comprises from 5 to 150 parts per million, based on a total weight of the ingredients in the pretreatment composition.

33. A method for treating a metal substrate comprising contacting the metal substrate with a pretreatment composition comprising a Group IIIB and/or Group IVB metal present in an amount of 50 ppm to 500 ppm based on a total weight of the ingredients in the pretreatment composition, free fluoride, lithium, and an electropositive metal;

wherein a molar ratio of the Group IIIB and/or Group IVB metal to the lithium is 100:1 to 1:10; and

wherein the composition is substantially free of phosphate ions.

34. The method according to claim 33, wherein the pretreatment composition further comprises molybdenum.

35. The method according to claim 33, further comprising electrophoretically depositing a coating composition onto the metal substrate.

36. The method according to claim 35, wherein the coating composition comprises yttrium.

37. A method of coating a metal substrate comprising electrophoretically depositing a coating composition onto the metal substrate, wherein the metal substrate comprises a surface layer, treated with the pretreatment composition of Claim 1.

38. A pretreated metal substrate comprising a surface layer, treated with the pretreatment composition of Claim 1.

39. An electrophoretically coated metal substrate comprising:

a treated surface layer comprising a film formed from a pretreatment composition comprising a Group IIIB and/or Group IVB metal present in an amount of 50 ppm to 500 ppm based on a total weight of the ingredients in the pretreatment composition, fluoride, lithium, and an electropositive metal on a surface of the metal substrate,

wherein a molar ratio of the Group IIIB and/or Group IVB metal to the lithium in the composition is 100:1 to 1:10, and wherein the composition is substantially free of phosphate ions; and

an electrophoretically deposited coating over at least a portion of the treated surface layer, wherein the coating comprises yttrium.

40. The method for treating a metal substrate according to claim 33, wherein the pretreatment composition is substantially free of resinous binder.

41. The method of coating a metal substrate according to claim 37, wherein the pretreatment composition is substantially free of resinous binder.

42. The pretreated metal substrate of claim 38, wherein the pretreatment composition is substantially free of resinous binder.

43. The electrophoretically coated metal substrate of claim 39, wherein the pretreatment composition is substantially free of resinous binder.