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(54) Title: LIQUID ELECTROPHOTOGRAPHIC INK COMPOSITIONS

(57) Abstract: A method of producing a material with a perovskite structure on a substrate by liquid electrophotographic printing comprises: either liquid electrophotographically printing onto the substrate a composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid; liquid electrophotographically printing onto the substrate a composition comprising a dispersion of a salt selected from BX2 and BX4 and a thermoplastic resin in a carrier liquid; and heat treating the printed compositions to form the material with a perovskite structure; or liquid electrophotographically printing onto the substrate a composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid to form the material with a perovskite structure; wherein A is a cation, B is a cation and X is an anion; wherein the thermoplastic resin comprises a copolymer of an alkylene monomer and a monomer having acidic side groups.



Liquid Electrophotographic Ink Compositions

Electrophotographic printing processes can involve creating an image on a photoconductive surface, applying an ink having charged particles to the photoconductive surface, such that they selectively bind to the image, and then
5 transferring the charged particles in the form of the image to a substrate.

The photoconductive surface may be on a cylinder and may be termed a photo imaging plate (PIP). The photoconductive surface is selectively charged with a latent electrophotographic image having image and background areas with different potentials. For example, an electrophotographic ink composition comprising charged
10 toner particles in a carrier liquid can be brought into contact with the selectively charged photoconductive surface. The charged toner particles adhere to the image areas of the latent image while the background areas remain clean. The image is then transferred to a substrate directly or, more commonly, by being first transferred to an intermediate transfer member, which can be a soft swelling blanket, and then to the
15 substrate.

Detailed Description

Before the present disclosure is disclosed and described, it is to be understood that this disclosure is not limited to the particular process steps and materials disclosed herein because such process steps and materials may vary somewhat. It is also to be
20 understood that the terminology used herein is used for the purpose of describing particular embodiments. The terms are not intended to be limiting because the scope is intended to be limited by the appended claims and equivalents thereof.

It is noted that, as used in this specification and the appended claims, the singular forms “a,” “an,” and “the” include plural referents unless the context clearly dictates
25 otherwise.

As used herein, “carrier fluid”, “carrier liquid,” “liquid carrier”, “carrier,” or “carrier vehicle” refers to the fluid in which pigment particles, resin, charge directors and other additives can be dispersed to form a liquid electrostatic ink composition or liquid electrophotographic ink composition. The carrier liquids may include a mixture of a
30 variety of different agents, such as surfactants, co-solvents, viscosity modifiers, and/or other possible ingredients.

As used herein, "liquid electrostatic ink composition" or "liquid electrophotographic composition" generally refers to an ink composition that is typically suitable for use in an electrostatic printing process, sometimes termed an electrophotographic printing process. It may comprise pigment particles having a thermoplastic resin thereon. The electrostatic ink composition may be a liquid electrostatic ink composition, in which the pigment particles having resin thereon are suspended in a carrier liquid. The pigment particles having resin thereon will typically be charged or capable of developing charge in an electric field, such that they display electrophoretic behaviour. A charge director may be present to impart a charge to the pigment particles having resin thereon.

As used herein, "co-polymer" refers to a polymer that is polymerized from at least two monomers.

As used herein, "melt flow rate" generally refers to the extrusion rate of a resin through an orifice of defined dimensions at a specified temperature and load, usually reported as temperature/load, e.g. 190°C/2.16 kg. Flow rates can be used to differentiate grades or provide a measure of degradation of a material as a result of molding. In the present disclosure, unless otherwise stated, "melt flow rate" is measured per ASTM D1238 Standard Test Method for Melt Flow Rates of Thermoplastics by Extrusion Plastometer, as known in the art. If a melt flow rate of a particular polymer is specified, unless otherwise stated, it is the melt flow rate for that polymer alone, in the absence of any of the other components of the liquid electrostatic ink composition.

As used herein, "acidity," "acid number," or "acid value" refers to the mass of potassium hydroxide (KOH) in milligrams that neutralizes one gram of a substance. The acidity of a polymer can be measured according to standard techniques, for example as described in ASTM D1386. If the acidity of a particular polymer is specified, unless otherwise stated, it is the acidity for that polymer alone, in the absence of any of the other components of the liquid toner composition.

As used herein, "melt viscosity" generally refers to the ratio of shear stress to shear rate at a given shear stress or shear rate. Testing is generally performed using a capillary rheometer. A plastic charge is heated in the rheometer barrel and is forced through a die with a plunger. The plunger is pushed either by a constant force or at constant rate depending on the equipment. Measurements are taken once the system has reached steady-state operation. One method used is measuring Brookfield viscosity @ 140°C, units are mPa·s or cPoise, as known in the art. Alternatively, the

melt viscosity can be measured using a rheometer, e.g. a commercially available AR-2000 Rheometer from Thermal Analysis Instruments, using the geometry of: 25mm steel plate-standard steel parallel plate, and finding the plate over plate rheometry isotherm at 120°C, 0.01 Hz shear rate. If the melt viscosity of a particular polymer is specified, unless otherwise stated, it is the melt viscosity for that polymer alone, in the absence of any of the other components of the electrostatic composition.

A certain monomer may be described herein as constituting a certain weight percentage of a polymer. This indicates that the repeating units formed from the said monomer in the polymer constitute said weight percentage of the polymer.

If a standard test is mentioned herein, unless otherwise stated, the version of the test to be referred to is the most recent at the time of filing this patent application.

As used herein, “electrostatic printing” or “electrophotographic printing” generally refers to the process that provides an image that is transferred from a photo imaging substrate either directly or indirectly via an intermediate transfer member to a print substrate, such as a plastic film. As such, the image is not substantially absorbed into the photo imaging substrate on which it is applied. Additionally, “electrophotographic printers” or “electrostatic printers” generally refer to those printers capable of performing electrophotographic printing or electrostatic printing, as described above. “Liquid electrostatic printing” is a specific type of electrostatic printing in which a liquid composition is employed in the electrophotographic process rather than a powder toner. An electrostatic printing process may involve subjecting the electrostatic composition to an electric field, for example, an electric field having a field gradient of 50-400 V/μm, or more, in some examples, 600-900V/μm, or more.

As used herein, “dispersion” generally refers to a composition comprising solid particles, for example, a thermoplastic resin, suspended in a liquid, wherein the solid particles are not soluble in the liquid at a temperature below 100°C at 10⁵ Pa. As used herein, a compound is not soluble in a liquid if a saturated solution comprises 5 wt.% or less of the compound, for example, 2 wt.% or less, 1 wt.% or less, 0.1 wt.% or less.

As used herein, “NVS” is an abbreviation of the term “non-volatile solids”.

As used herein, the term “about” is used to provide flexibility to a numerical range endpoint by providing that a given value may be a little above or a little below the endpoint to allow for variation in test methods or apparatus. The degree of flexibility of

this term can be dictated by the particular variable and would be within the knowledge of those skilled in the art to determine based on experience and the associated description herein.

As used herein, a plurality of items, structural elements, compositional elements, and/or materials may be presented in a common list for convenience. However, these lists should be construed as though each member of the list is individually identified as a separate and unique member. Thus, no individual member of such list should be construed as a de facto equivalent of any other member of the same list solely based on their presentation in a common group without indications to the contrary.

Concentrations, amounts, and other numerical data may be expressed or presented herein in a range format. It is to be understood that such a range format is used merely for convenience and brevity and thus should be interpreted flexibly to include not just the numerical values explicitly recited as the limits of the range, but also to include all the individual numerical values or sub-ranges encompassed within that range as if each numerical value and sub-range is explicitly recited. As an illustration, a numerical range of "about 1 wt% to about 5 wt%" should be interpreted to include not just the explicitly recited values of about 1 wt% to about 5 wt%, but also to include individual values and sub-ranges within the indicated range. Thus, included in this numerical range are individual values such as 2, 3.5, and 4 and sub-ranges such as from 1-3, from 2-4, and from 3-5, etc. This same principle applies to ranges reciting a single numerical value. Furthermore, such an interpretation should apply regardless of the breadth of the range or the characteristics being described.

As used herein, unless otherwise stated, wt.% values are to be taken as referring to a weight-for-weight (w/w) percentage of solids in the ink composition, and not including the weight of any carrier fluid present.

Unless otherwise stated, any feature described herein can be combined with any aspect or any other feature described herein.

In an aspect, there is provided a method of producing a material with a perovskite structure on a substrate wherein the material with a perovskite structure has a chemical formula selected from ABX_3 and A_2BX_6 .

The method of producing a material with a perovskite structure on a substrate by liquid electrophotographic (LEP) printing may comprise:

5

liquid electrophotographically printing onto the substrate a composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid;
liquid electrophotographically printing onto the substrate a composition comprising a dispersion of a salt BX₂ or a salt BX₄ and a thermoplastic resin in a
5 carrier liquid; and
heat treating the printed compositions to form the material with a perovskite structure;

wherein the material with a perovskite structure has a chemical formula selected from ABX₃ and A₂BX₆

10 wherein A is a cation, B is a cation and X is an anion; and

wherein the thermoplastic resin comprises a copolymer of an alkylene monomer and a monomer having acidic side groups.

The method of producing a material with a perovskite structure on a substrate by liquid electrophotographic printing may comprise:

15 liquid electrophotographically printing onto the substrate a composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid to form the material with a perovskite structure on the substrate;

wherein the material with a perovskite structure has a chemical formula selected from
20 ABX₃ and A₂BX₆

wherein A is a cation, and B is a cation and X is an anion; and

wherein the thermoplastic resin comprises a copolymer of an alkylene monomer and a monomer having acidic side groups.

In another aspect, there is provided an ink set. The ink set may comprise:

25 a liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid wherein the material with a perovskite structure has a chemical formula selected from ABX₃ and A₂BX₆;

or

30 a liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid; and

a liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ or a salt BX₄ and a thermoplastic resin in a carrier liquid;

wherein A is a cation, and B is a cation and X is an anion; and

wherein the thermoplastic resin comprises a copolymer of an alkylene monomer and a monomer having acidic side groups.

In a further aspect, there is provided a printed substrate. The printed substrate may comprise:

- 5 a substrate; and
 a liquid electrophotographically printed composition comprising a thermoplastic resin and a material with a perovskite structure disposed on the substrate;
wherein the material with a perovskite structure has a chemical formula selected from ABX_3 and A_2BX_6
- 10 wherein A is a cation, B is a cation and X is an anion; and
 wherein the thermoplastic resin comprises an alkylene monomer and a monomer having acidic side groups.

There is provided a method of producing a liquid electrophotographic ink composition, comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid, the

15 method comprising combining a salt AX, a thermoplastic resin and a carrier liquid;
wherein A is a cation and X is an anion; and wherein the thermoplastic resin comprises an alkylene monomer and a monomer having acidic side groups.

There is provided a method of producing a liquid electrophotographic ink composition comprising a dispersion of a salt BX_2 and a thermoplastic resin in a carrier liquid, the

20 method comprising combining a salt BX_2 and a thermoplastic resin in a carrier liquid,
wherein B is a cation and X is an anion; and wherein the thermoplastic resin comprises an alkylene monomer and a monomer having acidic side groups.

There is provided a method of producing a liquid electrophotographic ink composition comprising a dispersion of a salt BX_4 and a thermoplastic resin in a carrier liquid, the

25 method comprising combining a salt BX_4 and a thermoplastic resin in a carrier liquid,
wherein B is a cation and X is an anion; and wherein the thermoplastic resin comprises an alkylene monomer and a monomer having acidic side groups.

There is provided a method of producing a liquid electrophotographic ink composition comprising a material with a perovskite structure and a thermoplastic resin in a carrier

30 liquid, the method comprising:

- combining a salt AX, a salt selected from BX_2 and BX_4 , and a thermoplastic resin in a carrier liquid;
 or

combining a composition (for example, a liquid electrophotographic ink composition) comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid with a composition (for example, a liquid electrophotographic ink composition) comprising a dispersion of a salt selected from BX_2 and BX_4 and a thermoplastic resin in a carrier liquid.

The class of materials known as perovskites (i.e., materials with a perovskite crystal structure and a chemical formula ABX_3 or A_2BX_6) have been found to be particularly effective for use in photovoltaic cells and as semiconductor components in other electronic devices. Currently, many perovskites deteriorate on exposure to water and oxygen. Moreover, many current methods of forming perovskites on a substrate involve synthesis of the perovskite in solvents such as dimethylformamide (DMF) and dimethyl sulfoxide (DMSO), which are flammable and harmful to the environment.

Examples of the ink, printed substrate and method of producing a material with a perovskite structure described herein have been found to avoid or at least mitigate at least one of these difficulties. It has been found that perovskites disclosed herein can be prepared in a carrier liquid. Furthermore, it has been found that liquid electrophotographic printing can be used to form materials with a perovskite structure on substrates. Additionally, the printed substrates produced have been found to be more stable.

20 Ink Set

In an aspect, there is provided an ink set. The ink set may comprise a liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid; or a liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid and a liquid electrophotographic ink composition comprising a dispersion of a salt BX_2 or a salt BX_4 and a thermoplastic resin in a carrier liquid.

In some examples, the material with a perovskite structure is selected from materials with the chemical formula ABX_3 and materials with the chemical formula A_2BX_6 .

In some examples, A and B are each independently a cation and X is an anion. In some examples, the material with a perovskite structure has the chemical formula ABX_3 , wherein A is a monovalent cation, B is a divalent cation and X is a monovalent

anion. In some examples, the material with a perovskite structure has the chemical formula A_2BX_6 , wherein A is a monovalent cation, B is a tetravalent cation and X is a monovalent anion. In some examples, the material with a perovskite structure has the chemical formula A_2BX_6 , wherein A is a monovalent cation, B is a mixture of two
5 divalent cations and X is a monovalent anion.

In some examples, the thermoplastic resin comprises a copolymer of an alkylene monomer and a monomer having acidic side groups.

In some examples, the ink set may comprise a liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a
10 thermoplastic resin in a carrier liquid, wherein the material with a perovskite structure has a chemical formula selected from ABX_3 and A_2BX_6 .

In some examples, the ink set may comprise a liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid; and a liquid electrophotographic ink composition comprising a dispersion of a
15 salt BX_2 or a salt BX_4 and a thermoplastic resin in a carrier liquid. In some examples, the ink set may comprise a liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid; and a liquid electrophotographic ink composition comprising a dispersion of a salt BX_2 and a thermoplastic resin in a carrier liquid. In some examples, the ink set may comprise a
20 liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid; and a liquid electrophotographic ink composition comprising a dispersion of a salt BX_4 and a thermoplastic resin in a carrier liquid.

In some examples, the thermoplastic resin of each liquid electrophotographic ink composition in the ink set may be the same or different. In some examples, the
25 thermoplastic resin of each liquid electrophotographic ink composition in the ink set may be the same.

In some examples, the carrier liquid of each liquid electrophotographic ink composition in the ink set may be the same or different. In some examples, the carrier liquid of each liquid electrophotographic ink composition in the ink set may be the same.

30 In some examples, the ink set may comprise a liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid; and a liquid electrophotographic ink composition comprising a dispersion of a

salt BX_2 and a thermoplastic resin in a carrier liquid, wherein the two liquid electrophotographic ink compositions are present in amounts such that the ratio of salt AX to salt BX_2 is about 1:1 (by number of moles). In some examples, the ink set may comprise a liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid; and a liquid electrophotographic ink composition comprising a dispersion of a salt BX_4 and a thermoplastic resin in a carrier liquid, wherein the two liquid electrophotographic ink compositions are present in amounts such that the ratio of salt AX to salt BX_4 is about 2:1 (by number of moles).

Liquid electrophotographic Ink comprising AX

10 In some examples, the ink set comprises a liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid.

In some examples, the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid may comprise a carrier liquid and chargeable particles comprising a salt AX and a thermoplastic resin.

15 In some examples, the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid may further comprise a charge adjuvant.

In some examples, the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid may further
20 comprise a charge director. In some examples, the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid may further comprise a charge adjuvant and a charge director.

In some examples, the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid may further
25 comprise additives.

In some examples, the thermoplastic resin may constitute 5 wt.% or more of the total non-volatile solids of the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid, for example, 10 wt.% or more, 15 wt.% or more, 20 wt.% or more, 25 wt.% or more, 30 wt.% or more,
30 35 wt.% or more, 40 wt.% or more, 45 wt.% or more, 50 wt.% or more, 55 wt.% or more, 60 wt.% or more, 65 wt.% or more, 70 wt.% or more, or 75 wt.% or more of the total non-volatile solids of the liquid electrophotographic ink composition comprising a

dispersion of a salt AX and a thermoplastic resin in a carrier liquid. In some examples, the thermoplastic resin may constitute 75 wt.% or less of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid, for example, 70 wt.% or less, 65 wt.% or less, 60 wt.% or less, 55 wt.% or less, 50 wt.% or less, 45 wt.% or less, 40 wt.% or less, 35 wt.% or less, 30 wt.% or less, 25 wt.% or less, 20 wt.% or less, 15 wt.% or less, 10 wt.% or less, 5 wt.% or less of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid. In some examples, the thermoplastic resin may constitute 5 wt.% to 75 wt.% of the total non-volatile solids of the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid, for example, 10 wt.% to 70 wt.%, 15 wt.% to 65 wt.%, 20 wt.% to 60 wt.%, 25 wt.% to 55 wt.%, 30 wt.% to 50 wt.%, 35 wt.% to 70 wt.%, 40 wt.% to 50 wt.%, or 45 wt.% to 60 wt.% of the total non-volatile solids of the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid. In some examples, the thermoplastic resin may comprise about 46 wt.% of the total non-volatile solids of the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid.

In some examples, the salt AX may constitute 25 wt.% or more of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid, for example, 30 wt.% or more, 35 wt.% or more, 40 wt.% or more, 45 wt.% or more, 50 wt.% or more, 55 wt.% or more, 60 wt.% or more, 65 wt.% or more, 70 wt.% or more, 75 wt.% or more, 80 wt.% or more, 85 wt.% or more, 90 wt.% or more, or 95 wt.% or more of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid. In some examples, the salt AX may constitute 95 wt.% or less of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid, for example, 90 wt.% or less, 85 wt.% or less, 80 wt.% or less, 75 wt.% or less, 70 wt.% or less, 65 wt.% or less, 60 wt.% or less, 55 wt.% or less, 50 wt.% or less, 45 wt.% or less, 40 wt.% or less, 35 wt.% or less, 30 wt.% or less, or 25 wt.% or less of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid. In some examples, the salt AX may constitute 25 wt.% to 95 wt.% of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid.

liquid, for example, 30 wt.% to 90 wt.%, 35 wt.% to 85 wt.%, 40 wt.% to 80 wt.%, 45 wt.% to 75 wt.%, 50 wt.% to 70 wt.%, 35 wt.% to 65 wt.%, 45 wt.% to 60 wt.%, or 50 wt.% to 55 wt.% of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid. In some examples, the salt AX may comprise about 50 wt.% of the total non-volatile solids of the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid.

Liquid electrophotographic ink composition comprising BX₂

In some examples, the ink set comprises a liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid.

In some examples, the liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid may comprise a carrier liquid and chargeable particles comprising a salt BX₂ and a thermoplastic resin.

In some examples, the liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid may further comprise a charge adjuvant.

In some examples, the liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid may further comprise a charge director. In some examples, the liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid may further comprise a charge adjuvant and a charge director.

In some examples, the liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid may further comprise additives.

In some examples, the thermoplastic resin in the liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid may be the same as or different from the thermoplastic resin in the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid. In some examples, the thermoplastic resin in the liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid may be the same as the thermoplastic resin in the

liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid.

In some examples, the thermoplastic resin may constitute 5 wt.% or more of the total non-volatile solids of the liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid, for example, 10 wt.% or more, 15 wt.% or more, 20 wt.% or more, 25 wt.% or more, 30 wt.% or more, 35 wt.% or more, 40 wt.% or more, 45 wt.% or more, 50 wt.% or more, 55 wt.% or more, 60 wt.% or more, or 65 wt.% or more of the total non-volatile solids of the liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid. In some examples, the thermoplastic resin may constitute 65 wt.% or less of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid, for example, 60 wt.% or less, 55 wt.% or less, 50 wt.% or less, 45 wt.% or less, 40 wt.% or less, 35 wt.% or less, 30 wt.% or less, 25 wt.% or less, 20 wt.% or less, 15 wt.% or less, 10 wt.% or less, 5 wt.% or less of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid. In some examples, the thermoplastic resin may constitute 5 wt.% to 65 wt.% of the total non-volatile solids of the liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid, for example, 10 wt.% to 60 wt.%, 15 wt.% to 55 wt.%, 20 wt.% to 50 wt.%, 25 wt.% to 45 wt.%, 30 wt.% to 40 wt.%, or 35 wt.% to 40 wt.% of the total non-volatile solids of the liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid. In some examples, the thermoplastic resin may comprise about 36 wt.% of the total non-volatile solids of the liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid.

In some examples, the salt BX₂ may constitute 35 wt.% or more of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid, for example, 40 wt.% or more, 45 wt.% or more, 50 wt.% or more, 55 wt.% or more, 60 wt.% or more, 65 wt.% or more, 70 wt.% or more, 75 wt.% or more, 80 wt.% or more, 85 wt.% or more, 90 wt.% or more, 95 wt.% or more of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid. In some examples, the salt BX₂ may constitute 95 wt.% or less of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ and a

thermoplastic resin in a carrier liquid, for example, 90 wt.% or less, 85 wt.% or less, 80 wt.% or less, 75 wt.% or less, 70 wt.% or less, 65 wt.% or less, 60 wt.% or less, 55 wt.% or less, 50 wt.% or less, 45 wt.% or less, 40 wt.% or less, 35 wt.% or less of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a salt BX_2 and a thermoplastic resin in a carrier liquid. In some examples, the salt BX_2 may constitute 35 wt.% to 95 wt.% of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a salt BX_2 and a thermoplastic resin in a carrier liquid, for example, 40 wt.% to 90 wt.%, 45 wt.% to 85 wt.%, 50 wt.% to 80 wt.%, 55 wt.% to 75 wt.%, 60 wt.% to 70 wt.%, or 55 wt.% to 65 wt.% of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a salt BX_2 and a thermoplastic resin in a carrier liquid. In some examples, the salt BX_2 may comprise about 60 wt.% of the total non-volatile solids of the liquid electrophotographic ink composition comprising a dispersion of a salt BX_2 and a thermoplastic resin in a carrier liquid.

15 *Liquid electrophotographic ink composition comprising a salt BX_4*

In some examples, the ink set comprises a liquid electrophotographic ink composition comprising a dispersion of a salt BX_2 and a thermoplastic resin in a carrier liquid. In some examples, the liquid electrophotographic ink composition comprising a salt BX_4 may be as described above for the liquid electrophotographic ink composition comprising a salt BX_2 except that the salt BX_4 is used instead of the salt BX_2 .

Liquid electrophotographic ink composition comprising a material with a perovskite structure

In some examples, the ink set comprises a liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid, wherein the material with a perovskite structure has a chemical formula selected from ABX_3 and A_2BX_6 . In some examples, the material with a perovskite structure has the chemical formula ABX_3 . In some examples, the material with a perovskite structure has the chemical formula A_2BX_6 .

In some examples, the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid may comprise a carrier liquid and chargeable particles comprising a material with a perovskite structure and a thermoplastic resin.

In some examples, the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid may be producible by combining a liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid with a liquid electrophotographic ink composition comprising a dispersion of a salt BX₂ or a salt BX₄ and a thermoplastic resin in a carrier liquid.

In some examples, the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid may be producible by combining a salt AX, a salt selected from BX₂ and BX₄, a thermoplastic resin and a carrier liquid. In some examples, the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid, wherein the material with a perovskite structure has the chemical formula ABX₃, may be producible by combining a salt AX, a salt BX₂, a thermoplastic resin and a carrier liquid. In some examples, the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid, wherein the material with a perovskite structure has the chemical formula A₂BX₆, may be producible by combining a salt AX, a salt BX₄, a thermoplastic resin and a carrier liquid.

In some examples, the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid may further comprise a charge adjuvant.

In some examples, the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid may further comprise a charge director. In some examples, the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid may further comprise a charge adjuvant and a charge director.

In some examples, the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid may further comprise additives.

In some examples, the thermoplastic resin may constitute 5 wt.% or more of the total non-volatile solids of the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a

carrier liquid, for example, 10 wt.% or more, 15 wt.% or more, 20 wt.% or more, 25 wt.% or more, 30 wt.% or more, 35 wt.% or more, 40 wt.% or more, 45 wt.% or more, 50 wt.% or more, 55 wt.% or more, or 60 wt.% or more of the total non-volatile solids of the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid. In some examples, the thermoplastic resin may constitute 60 wt.% or less of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid, for example, 55 wt.% or less, 40 wt.% or less, 45 wt.% or less, 40 wt.% or less, 35 wt.% or less, 30 wt.% or less, 25 wt.% or less, 20 wt.% or less, 15 wt.% or less, 10 wt.% or less, or 5 wt.% or less of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid. In some examples, the thermoplastic resin may constitute 5 wt.% to 60 wt.% of the total non-volatile solids of the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid, for example, 10 wt.% to 55 wt.%, 15 wt.% to 50 wt.%, 20 wt.% to 45 wt.%, 25 wt.% to 45 wt.%, 30 wt.% to 60 wt.%, 35 wt.% to 55 wt.%, or 40 wt.% to 45 wt.% of the total non-volatile solids of the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid. In some examples, the thermoplastic resin may comprise about 44 wt.% of the total non-volatile solids of the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid.

In some examples, the material with a perovskite structure may constitute 20 wt.% or more of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid, for example, 20 wt.% or more, 25 wt.% or more, 30 wt.% or more, 35 wt.% or more, 40 wt.% or more, 45 wt.% or more, 50 wt.% or more, 55 wt.% or more, 60 wt.% or more, 65 wt.% or more, 70 wt.% or more, 75 wt.% or more, 80 wt.% or more, 85 wt.% or more, 90 wt.% or more, or 95 wt.% or more of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid. In some examples, the material with a perovskite structure may constitute 95 wt.% or less of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid, for example, 90

wt.% or less, 85 wt.% or less, 80 wt.% or less, 75 wt.% or less, 70 wt.% or less, 65 wt.% or less, 60 wt.% or less, 55 wt.% or less, 50 wt.% or less, 45 wt.% or less, or 40 wt.% or less of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid. In some examples, the material with a perovskite structure may constitute 40 wt.% to 95 wt.% of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid, for example, 45 wt.% to 90 wt.%, 50 wt.% to 85 wt.%, 45 wt.% to 80 wt.%, 50 wt.% to 75 wt.%, 45 wt.% to 70 wt.%, 50 wt.% to 65 wt.%, or 40 wt.% to 60 wt.% of the total solids of the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid. In some examples, the material with a perovskite structure may comprise about 52 wt.% of the total non-volatile solids of the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid.

Salt AX

In some examples, the salt AX may be a salt of a cation and an anion. In some examples, the salt AX may be a salt of a monovalent cation and a monovalent anion. In some examples, the salt AX may be a salt of a divalent cation and a divalent anion. In some examples, the salt AX may be a mixture of salts comprising monovalent cations and monovalent anions. In some examples, A is a monovalent cation or a mixture of monovalent cations. In some examples, X is a monovalent anion or a mixture of monovalent anions.

In some examples, A is selected from a metal cation, an organic cation or a mixture thereof. In some examples, A is selected from a monovalent metal cation, a monovalent organic cation, or a mixture thereof. In some examples, A is a monovalent metal cation or a mixture thereof. In some examples, A is a monovalent organic cation or a mixture thereof. In some examples, A is a mixture of a monovalent metal cation and a monovalent organic cation.

In some examples, A is an organic cation selected from primary aliphatic ammonium cations and primary aromatic ammonium cations. In some examples, A is selected from methylammonium (MA), formamidinium (FA), rubidium (Rb), caesium (Cs), and mixtures thereof. In some examples, A is caesium (Cs).

In some examples, X is a monovalent anion or a mixture of monovalent anions. In some examples, X is a halide ion. In some examples, X is selected from iodide, bromide, chloride and mixtures thereof. In some examples, X is selected from iodide, bromide and chloride. In some examples, X is bromide.

- 5 In some examples, AX may be selected from methylammonium iodide (MAI), methylammonium bromide (MABr), methylammonium chloride (MACl) formamidinium iodide (FAI), formamidinium bromide (FABr), formamidinium chloride (FABr), caesium iodide (Csl), caesium bromide (CsBr), caesium chloride (CsCl) rubidium iodide (RbI), rubidium bromide (RbBr), rubidium chloride (RbCl) or mixtures thereof. In some
10 examples, AX may be selected from methylammonium iodide (MAI), methylammonium bromide (MABr), formamidinium iodide (FAI), formamidinium bromide (FABr), caesium iodide (Csl), caesium bromide (CsBr), or mixtures thereof. In some examples, AX may be selected from methylammonium iodide (MAI), methylammonium bromide (MABr), formamidinium iodide (FAI), formamidinium bromide (FABr), caesium iodide (Csl) and
15 caesium bromide (CsBr). In some examples, AX may be selected from Csl and CsBr. In some examples, AX may be CsBr.

Salt BX₂

- In some examples, the salt BX₂ may be a salt of a cation and an anion. In some examples, the salt BX₂ may be a salt of a divalent cation and a monovalent anion. In
20 some examples, the salt BX₂ may be a salt of a tetravalent cation and a divalent anion. In some examples, the salt BX₂ may be a mixture of salts comprising divalent cations and monovalent anions. In some examples, B is a divalent cation or a mixture of divalent cations. In some examples, X is a monovalent anion or a mixture of monovalent anions. In some examples, X in BX₂ may be different from X in AX. In
25 some examples, X in BX₂ may be the same as X in AX.

In some examples, B is a divalent metal cation or a mixture of divalent metal cations. In some examples, B is a divalent metal cation.

- In some examples, B is selected from lead (Pb), germanium (Ge), tin (Sn), antimony (Sb), bismuth (Bi), copper (Cu), manganese (Mn²⁺), cobalt (Co²⁺) and mixtures thereof.
30 In some examples, B is selected from lead (Pb), germanium (Ge), tin (Sn), antimony (Sb), bismuth (Bi), Copper (Cu) and mixtures thereof.

In some examples, X is as described above for AX.

In some examples, BX_2 may be selected from SnI_2 , $SnBr_2$, $SnCl_2$, PbI_2 , $PbBr_2$, $PbCl_2$ and combinations thereof. In some examples, BX_2 may be selected from SnI_2 , $SnBr_2$, PbI_2 , $PbBr_2$ and combinations thereof. In some examples, BX_2 may be selected from SnI_2 , $SnBr_2$, PbI_2 and $PbBr_2$. In some examples, BX_2 may be selected from SnI_2 and $SnBr_2$. In some examples, BX_2 may be $SnBr_2$.

Salt BX_4

In some examples, the salt BX_4 may be a salt of a cation and an anion. In some examples, the salt BX_4 may be a salt of a tetravalent cation and a monovalent anion. In some examples, the salt BX_4 may be a mixture of salts comprising tetravalent cations and monovalent anions. In some examples, B is a tetravalent cation or a mixture of tetravalent cations. In some examples, X is a monovalent anion or a mixture of monovalent anions. In some examples, X in BX_2 may be the same as X in AX.

In some examples, B is a tetravalent metal cation or a mixture of tetravalent metal cations. In some examples, B is a tetravalent metal cation. In some examples, B is Sn^{4+} .

In some examples, BX_4 is selected from SnI_4 , $SnBr_4$, $SnCl_4$ and combinations thereof.

Material with a perovskite structure

In some examples, the material with a perovskite structure may be a perovskite wherein A is a cation or a mixture of cations, B is a cation or a mixture of cations and X is an anion or a mixture of anions. In some examples, the material with a perovskite structure may have the chemical formula ABX_3 wherein A is a monovalent cation or a mixture of monovalent cations, B is a divalent cation or a mixture of divalent cations and X is a monovalent anion or a mixture of monovalent anions. In some examples, the material with a perovskite structure may have the chemical formula ABX_3 wherein A is a divalent cation or a mixture of divalent cations, B is a tetravalent cation or a mixture of tetravalent cations and X is a divalent anion or a mixture of divalent anions. In some examples, the material with a perovskite structure may have the chemical formula A_2BX_6 wherein A is a monovalent cation or a mixture of monovalent cations, B is a tetravalent cation or mixture of tetravalent cations and X is a monovalent anion or a mixture of monovalent anions. In some examples, the material with a perovskite structure may have the chemical formula ABX_3 wherein A is a monovalent cation, B is a divalent cation and X is a monovalent anion or a mixture of monovalent anions. In

some examples, the material with a perovskite structure may have the chemical formula ABX_3 wherein A is a monovalent cation, B is a divalent cation and X is a monovalent anion.

In some examples, A is as described above for AX. In some examples, B is as described above for BX_2 or as described above for BX_4 . In some examples, X is as described above for AX or BX_2 or BX_4 .

As used herein, the term perovskite does not specifically refer to the perovskite mineral, $CaTiO_3$ but instead refers to any material that has the same type of crystal structure as calcium titanium oxide. As used herein, the term perovskite structure indicates that the material has the perovskite type crystal structure.

In some examples, the material with a perovskite structure may be selected from $CsSnBr_3$, Cs_2SnBr_6 , $Rb_z[Cs_y(MA_xFA_{1-x})_{1-y}]_{1-z}Pb_1M_{1-l}(I_{1-n-m}Br_mCl_n)_3$ (wherein $0 \leq x \leq 1$, $0 \leq y \leq 1$, $0 \leq z \leq 1$, $0 \leq l \leq 1$, $0 \leq m \leq 1$, $0 \leq n \leq 1$; $M=Sn$ or In), and $Rb_z[Cs_y(MA_xFA_{1-x})_{1-y}]_{1-z}B(I_{1-n-m}Br_mCl_n)_3$ (wherein $0 \leq x \leq 1$, $0 \leq y \leq 1$, $0 \leq z \leq 1$, $0 \leq m \leq 1$, $0 \leq n \leq 1$; $B=Ge$, Sn , Sb , Bi or Cu). In some examples, the material with a perovskite structure may be $CsSnBr_3$.

Thermoplastic Resin

In some examples, the thermoplastic resin may comprise a copolymer of an alkylene monomer and a monomer having acidic side groups. In some examples, the alkylene monomer may be selected from ethylene and propylene. In some examples, the alkylene monomer may be ethylene. In some examples, the monomer having acidic side groups may be selected from acrylic acid and methacrylic acid.

In some examples, the thermoplastic resin comprises a copolymer of ethylene and a monomer selected from acrylic acid and methacrylic acid.

The thermoplastic resin may be referred to herein as a resin.

In some examples, the liquid electrophotographic ink composition comprises chargeable particles (i.e., having or capable of developing a charge, for example, in an electromagnetic field) including the thermoplastic resin. In some examples, the chargeable particles may comprise the salt AX, the salt BX_2 , the salt BX_4 or the material with a perovskite structure.

In some examples, the thermoplastic resin may comprise a polymer selected from ethylene acrylic acid copolymers; ethylene methacrylic acid copolymers; ethylene vinyl acetate copolymers; copolymers of ethylene (e.g. 80 wt.% to 99.9 wt.%), and alkyl (e.g. C1 to C5) ester of methacrylic or acrylic acid (e.g. 0.1 wt.% to 20 wt.%); copolymers of ethylene (e.g. 80 wt.% to 99.9 wt.%), acrylic or methacrylic acid (e.g. 0.1 wt.% to 20 wt.%) and alkyl (e.g. C1 to C5) ester of methacrylic or acrylic acid (e.g. 0.1 wt.% to 20 wt.%)

5 polyethylene; polystyrene; isotactic polypropylene (crystalline); ethylene ethyl acrylate; polyesters; polyvinyl toluene; polyamides; styrene/butadiene copolymers; epoxy resins; acrylic resins (e.g. copolymer of acrylic or methacrylic acid and at least one alkyl ester of acrylic or methacrylic acid wherein alkyl is, in some examples, from 1 to about 20 carbon atoms, such as methyl methacrylate (e.g. 50 wt.% to 90 wt.%)

10 /methacrylic acid (e.g. 0 wt.% to 20 wt.%) /ethylhexylacrylate (e.g. 10 wt.% to 50 wt.%)

15 or glycidyl methacrylate (GMA) terpolymers; ethylene-acrylic acid ionomers and combinations thereof.

The polymer having acidic side groups may have an acidity of 50 mg KOH/g or more, in some examples an acidity of 60 mg KOH/g or more, in some examples an acidity of 70 mg KOH/g or more, in some examples an acidity of 80 mg KOH/g or more, in some examples an acidity of 90 mg KOH/g or more, in some examples an acidity of 100 mg KOH/g or more, in some examples an acidity of 105 mg KOH/g or more, in some examples 110 mg KOH/g or more, in some examples 115 mg KOH/g or more. The polymer having acidic side groups may have an acidity of 200 mg KOH/g or less, in some examples 190 mg or less, in some examples 180 mg or less, in some examples 130 mg KOH/g or less, in some examples 120 mg KOH/g or less. Acidity of a polymer, as measured in mg KOH/g, can be measured using standard procedures known in the art, for example, using the procedure described in ASTM D1386.

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The thermoplastic resin may comprise a polymer having acidic side groups that has a melt flow rate of less than about 60 g/10 minutes, in some examples about 50 g/10 minutes or less, in some examples about 40 g/10 minutes or less, in some examples 30 g/10 minutes or less, in some examples 20 g/10 minutes or less, in some examples 10 g/10 minutes or less. In some examples, all polymers having acidic side groups and/or ester groups in the particles each individually have a melt flow rate of less than 90 g/10 minutes, 80 g/10 minutes or less, in some examples 70 g/10 minutes or less, in some examples 60 g/10 minutes or less.

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The polymer having acidic side groups can have a melt flow rate of about 10 g/10 minutes to about 120 g/10 minutes, in some examples about 10 g/10 minutes to about 70 g/10 minutes, in some examples about 10 g/10 minutes to 40 g/10 minutes, in some examples 20 g/10 minutes to 30 g/10 minutes. The polymer having acidic side groups
5 can have a melt flow rate of in some examples about 50 g/10 minutes to about 120 g/10 minutes, in some examples 60 g/10 minutes to about 100 g/10 minutes. The melt flow rate can be measured using standard procedures known in the art, for example, as described in ASTM D1238.

The thermoplastic resin may comprise a copolymer of an alkylene monomer and a
10 monomer having acidic side groups. In some examples, the alkylene monomer may be selected from ethylene and propylene. In some examples, the monomer having acidic side groups may be selected from methacrylic acid and acrylic acid. In some examples, the thermoplastic resin may comprise a copolymer of an alkylene monomer and a monomer selected from methacrylic acid and acrylic acid. In some examples, the
15 thermoplastic resin may comprise a copolymer of ethylene and a monomer selected from methacrylic acid and acrylic acid.

In some examples, the polymer having acidic side groups is a copolymer of an alkylene monomer and a monomer selected from acrylic acid and methacrylic acid. In some examples, the thermoplastic resin may comprise a copolymer of an alkylene monomer
20 and a monomer selected from acrylic acid and methacrylic acid.

The acidic side groups may be in free acid form or may be in the form of an anion and associated with one or more counterions, typically metal counterions, e.g. a metal selected from the alkali metals, such as lithium, sodium and potassium, alkali earth metals, such as magnesium or calcium, and transition metals, such as zinc. The
25 polymer having acidic side groups can be selected from resins such as copolymers of ethylene and an ethylenically unsaturated acid of either acrylic acid or methacrylic acid; and ionomers thereof, such as methacrylic acid and ethylene-acrylic or methacrylic acid copolymers which are at least partially neutralized with metal ions (e.g. Zn, Na, Li) such as SURLYN® ionomers. The polymer comprising acidic side groups can be a
30 copolymer of ethylene and an ethylenically unsaturated acid of either acrylic or methacrylic acid, where the ethylenically unsaturated acid of either acrylic or methacrylic acid constitute from 5 wt.% to about 25 wt.% of the copolymer, in some examples from 10 wt.% to about 20 wt.% of the copolymer.

The thermoplastic resin may comprise two different polymers having acidic side groups. The two polymers having acidic side groups may have different acidities, which may fall within the ranges mentioned above. The thermoplastic resin may comprise a first polymer having acidic side groups that has an acidity of from 50 mg KOH/g to 110 mg KOH/g and a second polymer having acidic side groups that has an acidity of 110 mg KOH/g to 130 mg KOH/g.

The resin may comprise two different polymers having acidic side groups: a first polymer having acidic side groups that has a melt flow rate of about 10 g/10 minutes to about 50 g/10 minutes and an acidity of from 50 mg KOH/g to 110 mg KOH/g, and a second polymer having acidic side groups that has a melt flow rate of about 50 g/10 minutes to about 120 g/10 minutes and an acidity of 110 mg KOH/g to 130 mg KOH/g. The first and second polymers may be absent of ester groups.

The resin may comprise a copolymer of ethylene and acrylic acid and a copolymer of ethylene and methacrylic acid.

The resin may comprise two different polymers having acidic side groups: a first polymer that is a copolymer of ethylene (e.g. 92 to 85 wt.%, in some examples about 89 wt.%) and acrylic or methacrylic acid (e.g. 8 to 15 wt.%, in some examples about 11 wt.%) having a melt flow rate of 80 to 110 g/10 minutes and a second polymer that is a copolymer of ethylene (e.g. about 80 to 92 wt.%, in some examples about 85 wt.%) and acrylic acid (e.g. about 18 to 12 wt.%, in some examples about 15 wt.%), having a melt viscosity lower than that of the first polymer, the second polymer for example having a melt viscosity of 15000 poise or less, in some examples a melt viscosity of 10000 poise or less, in some examples 1000 poise or less, in some examples 100 poise or less, in some examples 50 poise or less, in some examples 10 poise or less. Melt viscosity can be measured using standard techniques. The melt viscosity can be measured using a rheometer, e.g. a commercially available AR-2000 Rheometer from Thermal Analysis Instruments, using the geometry of: 25mm steel plate-standard steel parallel plate, and finding the plate over plate rheometry isotherm at 120°C, 0.01 Hz shear rate.

In any of the resins mentioned above, the ratio of the first polymer having acidic side groups to the second polymer having acidic side groups can be from about 10:1 to about 2:1. In another example, the ratio can be from about 6:1 to about 3:1, in some examples about 4:1.

The resin may comprise a polymer having a melt viscosity of 15000 poise or less, in some examples a melt viscosity of 10000 poise or less, in some examples 1000 poise or less, in some examples 100 poise or less, in some examples 50 poise or less, in some examples 10 poise or less; said polymer may be a polymer having acidic side groups as described herein. The resin may comprise a first polymer having a melt viscosity of 15000 poise or more, in some examples 20000 poise or more, in some examples 50000 poise or more, in some examples 70000 poise or more; and in some examples, the resin may comprise a second polymer having a melt viscosity less than the first polymer, in some examples a melt viscosity of 15000 poise or less, in some examples a melt viscosity of 10000 poise or less, in some examples 1000 poise or less, in some examples 100 poise or less, in some examples 50 poise or less, in some examples 10 poise or less. The resin may comprise a first polymer having a melt viscosity of more than 60000 poise, in some examples from 60000 poise to 100000 poise, in some examples from 65000 poise to 85000 poise; a second polymer having a melt viscosity of from 15000 poise to 40000 poise, in some examples 20000 poise to 30000 poise, and a third polymer having a melt viscosity of 15000 poise or less, in some examples a melt viscosity of 10000 poise or less, in some examples 1000 poise or less, in some examples 100 poise or less, in some examples 50 poise or less, in some examples 10 poise or less; an example of the first polymer is Nucrel 960 (from DuPont), an example of the second polymer is Nucrel 699 (from DuPont), and an example of the third polymer is AC-5120 (from Honeywell). In some examples, the resin may comprise a first polymer having a melt viscosity of from 15000 poise to 40000 poise, in some examples 20000 poise to 30000 poise, and a second polymer having a melt viscosity of 15000 poise or less, in some examples a melt viscosity of 10000 poise or less, in some examples 1000 poise or less, in some examples 100 poise or less, in some examples 50 poise or less, in some examples 10 poise or less; an example of the first polymer is Nucrel 699 (from DuPont), and an example of the second polymer is AC-5120 (from Honeywell). The first, second and third polymers may be polymers having acidic side groups as described herein. The melt viscosity can be measured using a rheometer, e.g. a commercially available AR-2000 Rheometer from Thermal Analysis Instruments, using the geometry of: 25 mm steel plate-standard steel parallel plate, and finding the plate over plate rheometry isotherm at 120°C, 0.01 Hz shear rate.

If the resin comprises a single type of resin polymer, the resin polymer (excluding any other components of the electrostatic ink composition) may have a melt viscosity of

6000 poise or more, in some examples a melt viscosity of 8000 poise or more, in some examples a melt viscosity of 10000 poise or more, in some examples a melt viscosity of 12000 poise or more. If the resin comprises a plurality of polymers all the polymers of the resin may together form a mixture (excluding any other components of the electrostatic ink composition) that has a melt viscosity of 6000 poise or more, in some examples a melt viscosity of 8000 poise or more, in some examples a melt viscosity of 10000 poise or more, in some examples a melt viscosity of 12000 poise or more. Melt viscosity can be measured using standard techniques. The melt viscosity can be measured using a rheometer, e.g. a commercially available AR-2000 Rheometer from Thermal Analysis Instruments, using the geometry of: 25mm steel plate-standard steel parallel plate, and finding the plate over plate rheometry isotherm at 120°C, 0.01 Hz shear rate.

The resin may comprise two different polymers having acidic side groups that are selected from copolymers of ethylene and an ethylenically unsaturated acid of either methacrylic acid or acrylic acid; and ionomers thereof, such as methacrylic acid and ethylene-acrylic or methacrylic acid copolymers which are at least partially neutralized with metal ions (e.g. Zn, Na, Li) such as SURLYN® ionomers.

The resin may comprise (i) a first polymer that is a copolymer of ethylene and an ethylenically unsaturated acid of either acrylic acid and methacrylic acid, wherein the ethylenically unsaturated acid of either acrylic or methacrylic acid constitutes from 8 wt.% to about 16 wt.% of the copolymer, in some examples 10 wt.% to 16 wt.% of the copolymer; and (ii) a second polymer that is a copolymer of ethylene and an ethylenically unsaturated acid of either acrylic acid and methacrylic acid, wherein the ethylenically unsaturated acid of either acrylic or methacrylic acid constitutes from 12 wt.% to about 30 wt.% of the copolymer, in some examples from 14 wt.% to about 20 wt.% of the copolymer, in some examples from 16 wt.% to about 20 wt.% of the copolymer in some examples from 17 wt.% to 19 wt.% of the copolymer.

In an example, the resin constitutes about 5 to 90%, in some examples about 5 to 80% by weight of the total solids of the electrostatic ink composition. In another example, the resin constitutes about 10 to 60% by weight of the total solids of the electrostatic ink composition. In another example, the resin constitutes about 15 to 40% by weight of the total solids of the electrostatic ink composition. In another example, the resin constitutes about 60 to 95% by weight, in some examples, from 65 to 90% by weight, from 65 to 80% by weight of the total solids of the electrostatic ink composition.

The resin may comprise a polymer having acidic side groups, as described above (which may be free of ester side groups), and a polymer having ester side groups. The polymer having ester side groups is, in some examples, a thermoplastic polymer. The polymer having ester side groups may further comprise acidic side groups. The
5 polymer having ester side groups may be a copolymer of a monomer having ester side groups and a monomer having acidic side groups. The polymer may be a copolymer of a monomer having ester side groups, a monomer having acidic side groups, and a monomer absent of any acidic and ester side groups. The monomer having ester side groups may be a monomer selected from esterified acrylic acid or esterified methacrylic
10 acid. The monomer having acidic side groups may be a monomer selected from acrylic or methacrylic acid. The monomer absent of any acidic and ester side groups may be an alkylene monomer, including, but not limited to, ethylene or propylene. The esterified acrylic acid or esterified methacrylic acid may, respectively, be an alkyl ester of acrylic acid or an alkyl ester of methacrylic acid. The alkyl group in the alkyl ester of
15 acrylic or methacrylic acid may be an alkyl group having 1 to 30 carbons, in some examples 1 to 20 carbons, in some examples 1 to 10 carbons; in some examples selected from methyl, ethyl, iso-propyl, n-propyl, t-butyl, iso-butyl, n-butyl and pentyl.

The polymer having ester side groups may be a copolymer of a first monomer having ester side groups, a second monomer having acidic side groups and a third monomer
20 which is an alkylene monomer absent of any acidic and ester side groups. The polymer having ester side groups may be a copolymer of (i) a first monomer having ester side groups selected from esterified acrylic acid or esterified methacrylic acid, in some examples an alkyl ester of acrylic or methacrylic acid, (ii) a second monomer having acidic side groups selected from acrylic or methacrylic acid and (iii) a third monomer
25 which is an alkylene monomer selected from ethylene and propylene. The first monomer may constitute 1 to 50% by weight of the copolymer, in some examples 5 to 40% by weight, in some examples 5 to 20% by weight of the copolymer, in some examples 5 to 15% by weight of the copolymer. The second monomer may constitute 1 to 50% by weight of the copolymer, in some examples 5 to 40% by weight of the
30 copolymer, in some examples 5 to 20% by weight of the copolymer, in some examples 5 to 15% by weight of the copolymer. In an example, the first monomer constitutes 5 to 40% by weight of the copolymer, the second monomer constitutes 5 to 40% by weight of the copolymer, and with the third monomer constituting the remaining weight of the copolymer. In an example, the first monomer constitutes 5 to 15% by weight of the
35 copolymer, the second monomer constitutes 5 to 15% by weight of the copolymer, with

the third monomer constituting the remaining weight of the copolymer. In an example, the first monomer constitutes 8 to 12% by weight of the copolymer, the second monomer constitutes 8 to 12% by weight of the copolymer, with the third monomer constituting the remaining weight of the copolymer. In an example, the first monomer constitutes about 10% by weight of the copolymer, the second monomer constitutes about 10% by weight of the copolymer, and with the third monomer constituting the remaining weight of the copolymer. The polymer having ester side groups may be selected from the Bynel ® class of monomer, including Bynel 2022 and Bynel 2002, which are available from DuPont ®.

- 10 The polymer having ester side groups may constitute 1% or more by weight of the total amount of the resin polymers in the resin, e.g. the total amount of the polymer or polymers having acidic side groups and polymer having ester side groups. The polymer having ester side groups may constitute 5% or more by weight of the total amount of the resin polymers in the resin, in some examples 8% or more by weight of the total amount of the resin polymers in the resin, in some examples 10% or more by weight of the total amount of the resin polymers in the resin, in some examples 15% or more by weight of the total amount of the resin polymers in the resin, in some examples 20% or more by weight of the total amount of the resin polymers in the resin, in some examples 25% or more by weight of the total amount of the resin polymers in the resin, in some examples 30% or more by weight of the total amount of the resin polymers in the resin, in some examples 35% or more by weight of the total amount of the resin polymers in the resin. The polymer having ester side groups may constitute from 5% to 50% by weight of the total amount of the resin polymers in the resin, in some examples 10% to 40% by weight of the total amount of the resin polymers in the resin, in some examples 15% to 30% by weight of the total amount of the polymers in the resin.

- 30 The polymer having ester side groups may have an acidity of 50 mg KOH/g or more, in some examples an acidity of 60 mg KOH/g or more, in some examples an acidity of 70 mg KOH/g or more, in some examples an acidity of 80 mg KOH/g or more. The polymer having ester side groups may have an acidity of 100 mg KOH/g or less, in some examples 90 mg KOH/g or less. The polymer having ester side groups may have an acidity of 60 mg KOH/g to 90 mg KOH/g, in some examples 70 mg KOH/g to 80 mg KOH/g.

The polymer having ester side groups may have a melt flow rate of about 10 g/10 minutes to about 120 g/10 minutes, in some examples about 10 g/10 minutes to about

50 g/10 minutes, in some examples about 20 g/10 minutes to about 40 g/10 minutes, in some examples about 25 g/10 minutes to about 35 g/10 minutes.

In an example, the polymer or polymers of the resin can be selected from the Nucrel family of toners (e.g. Nucrel 403™, Nucrel 407™, Nucrel 609HS™, Nucrel 908HS™, Nucrel 1202HC™, Nucrel 30707™, Nucrel 1214™, Nucrel 903™, Nucrel 3990™, Nucrel 910™, Nucrel 925™, Nucrel 699™, Nucrel 599™, Nucrel 960™, Nucrel RX 76™, Nucrel 2806™, Bynell 2002, Bynell 2014, and Bynell 2020 (sold by E. I. du PONT)), the Aclyn family of toners (e.g. Aclyn 201, Aclyn 246, Aclyn 285, and Aclyn 295), AC-5120 and AC 580 (sold by Honeywell), and the Lotader family of toners (e.g. Lotader 2210, Lotader, 3430, and Lotader 8200 (sold by Arkema)).

In some examples, the resin may constitute 5% to 99% by weight of the total solids in the liquid electrophotographic ink composition, in some examples 50% to 90% by weight of the total solids of the liquid electrophotographic ink composition, in some examples 65% to 80% by weight of the total solids of the liquid electrophotographic ink composition.

Carrier Liquid

In some examples, when printing, the liquid electrophotographic ink composition comprises a carrier liquid. Generally, the carrier liquid can act as a dispersing medium for the other components in the liquid electrophotographic ink composition. For example, the carrier liquid can comprise or be a hydrocarbon, silicone oil, vegetable oil, and so forth.

The carrier liquid can include, but is not limited to, an insulating, non-polar, non-aqueous liquid that can be used as a medium for toner particles. The carrier liquid can include compounds that have a resistivity in excess of about 10^9 ohm·cm. The carrier liquid may have a dielectric constant below about 5, in some examples below about 3. The carrier liquid can include, but is not limited to, hydrocarbons. The hydrocarbon can include, but is not limited to, an aliphatic hydrocarbon, an isomerized aliphatic hydrocarbon, branched chain aliphatic hydrocarbons, aromatic hydrocarbons, and combinations thereof. Examples of the carrier liquids include, but are not limited to, aliphatic hydrocarbons, isoparaffinic compounds, paraffinic compounds, dearomatized hydrocarbon compounds, and the like.

In some examples, the carrier liquid may be a hydrocarbon. In some examples, the carrier liquid may be a branched chain hydrocarbon. In some examples, the branched chain hydrocarbon may comprise 5 to 15 carbon atoms, for example, 10 to 15 carbon atoms, or 11 to 12 carbon atoms. In some examples, the carrier liquid may be selected
5 from liquids comprising a mixture of branched chain hydrocarbons having 5 to 15 carbon atoms, for example, 10 to 15 carbon atoms or 11 to 12 carbon atoms.

In particular, the liquid carriers can include, but are not limited to, Isopar-G™, Isopar-H™, Isopar-L™, Isopar-M™, Isopar-K™, Isopar-V™, Norpar 12™, Norpar 13™, Norpar 15™, Exxol D40™, Exxol D80™, Exxol D100™, Exxol D130™, and Exxol
10 D140™ (each sold by EXXON CORPORATION); Teclen N-16™, Teclen N-20™, Teclen N-22™, Nisseki Naphthesol L™, Nisseki Naphthesol M™, Nisseki Naphthesol H™, #0 Solvent L™, #0 Solvent M™, #0 Solvent H™, Nisseki Isosol 300™, Nisseki Isosol 400™, AF-4™, AF-5™, AF-6™ and AF-7™ (each sold by NIPPON OIL CORPORATION); IP Solvent 1620™ and IP Solvent 2028™ (each sold by IDEMITSU
15 PETROCHEMICAL CO., LTD.); Amsco OMS™ and Amsco 460™ (each sold by AMERICAN MINERAL SPIRITS CORP.); and Electron, Positron, New II, Purogen HF (100% synthetic terpenes) (sold by ECOLINK™).

Before liquid electrophotographic printing, the carrier liquid can constitute about 20% to 99.5% by weight of the liquid electrostatic ink composition, in some examples 50% to
20 99.5% by weight of the liquid electrostatic ink composition. Before printing, the carrier liquid may constitute about 40% to 90% by weight of the liquid electrostatic ink composition. Before printing, the carrier liquid may constitute about 60% to 80% by weight of the liquid electrostatic ink composition. Before printing, the liquid carrier may constitute about 90% to 99.5% by weight of the liquid electrostatic ink composition, in
25 some examples 95% to 99% by weight of the liquid electrostatic ink composition.

The liquid electrostatic ink composition, once electrostatically printed on the substrate, may be substantially free from liquid carrier. In an electrostatic printing process and/or afterwards, the liquid carrier may be removed, for example, by an electrophoresis processes during printing and/or evaporation, such that substantially just solids are
30 transferred to the substrate. Substantially free from liquid carrier may indicate that liquid electrostatically printed ink contains less than 5 wt.% liquid carrier, in some examples, less than 2 wt.% liquid carrier, in some examples less than 1 wt.% liquid carrier, in some examples less than 0.5 wt.% liquid carrier. In some examples, liquid electrostatically printed ink is free from liquid carrier.

Charge director

In some examples, the LEP ink composition further includes a charge director. The charge director may be added in order to impart and/or maintain sufficient electrostatic charge on the ink particles, which may be particles comprising the thermoplastic resin.

- 5 In some examples, the charge director may comprise ionic compounds, particularly metal salts of fatty acids, metal salts of sulfo-succinates, metal salts of oxyphosphates, metal salts of alkyl-benzenesulfonic acid, metal salts of aromatic carboxylic acids or sulfonic acids, as well as zwitterionic and non-ionic compounds, such as polyoxyethylated alkylamines, lecithin, polyvinylpyrrolidone, organic acid esters of
- 10 polyvalent alcohols, etc. The charge director can be selected from, but is not limited to, oil-soluble petroleum sulfonates (e.g. neutral Calcium Petronate™, neutral Barium Petronate™, and basic Barium Petronate™), polybutylene succinimides (e.g. OLOA™ 1200 and Amoco 575), and glyceride salts (e.g. sodium salts of phosphated mono- and diglycerides with unsaturated and saturated acid substituents), sulfonic acid salts
- 15 including, but not limited to, barium, sodium, calcium, and aluminum salts of sulfonic acid. The sulfonic acids may include, but are not limited to, alkyl sulfonic acids, aryl sulfonic acids, and sulfonic acids of alkyl succinates. The charge director can impart a negative charge or a positive charge on the resin-containing particles of a LEP ink composition.
- 20 In some examples, the liquid electrostatic ink composition comprises a charge director comprising a simple salt. The ions constructing the simple salts are all hydrophilic. The simple salt may include a cation selected from the group consisting of Mg, Ca, Ba, NH₄, *tert*-butyl ammonium, Li⁺, and Al³⁺, or from any sub-group thereof. The simple salt may include an anion selected from the group consisting of SO₄²⁻, PO₃⁻, NO₃⁻, HPO₄²⁻, CO₃²⁻, acetate, trifluoroacetate (TFA), Cl⁻, BF₄⁻, F⁻, ClO₄⁻, and TiO₃⁴⁻ or from any sub-group thereof. The simple salt may be selected from CaCO₃, Ba₂TiO₃, Al₂(SO₄), Al(NO₃)₃, Ca₃(PO₄)₂, BaSO₄, BaHPO₄, Ba₂(PO₄)₃, CaSO₄, (NH₄)₂CO₃, (NH₄)₂SO₄, NH₄OAc, *tert*-butyl ammonium bromide, NH₄NO₃, LiTFA, Al₂(SO₄)₃, LiClO₄ and LiBF₄, or any sub-group thereof.
- 25
- 30 In some examples, the electrostatic ink composition comprises a charge director comprising a sulfosuccinate salt of the general formula MA_n, wherein M is a metal, n is the valence of M, and A is an ion of the general formula (I): [R¹-O-C(O)CH₂CH(SO₃-)-C(O)-O-R²], wherein each of R¹ and R² is an alkyl group. In some examples each of R¹ and R² is an aliphatic alkyl group. In some examples, each of R¹ and R² independently

is a C6-25 alkyl. In some examples, said aliphatic alkyl group is linear. In some examples, said aliphatic alkyl group is branched. In some examples, said aliphatic alkyl group includes a linear chain of more than 6 carbon atoms. In some examples, R¹ and R² are the same. In some examples, at least one of R¹ and R² is C₁₃H₂₇. In some
5 examples, M is Na, K, Cs, Ca, or Ba.

In some examples, the charge director comprises at least one micelle forming salt and nanoparticles of a simple salt as described above. The simple salts are salts that do not form micelles by themselves, although they may form a core for micelles with a micelle forming salt. The sulfosuccinate salt of the general formula MA_n is an example
10 of a micelle forming salt. The charge director may be substantially free of an acid of the general formula HA, where A is as described above. The charge director may include micelles of said sulfosuccinate salt enclosing at least some of the nanoparticles of the simple salt. The charge director may include at least some nanoparticles of the simple salt having a size of 200 nm or less, and/or in some examples 2 nm or more.

15 The charge director may include one of, some of or all of (i) soya lecithin, (ii) a barium sulfonate salt, such as basic barium petronate (BBP), and (iii) an isopropyl amine sulfonate salt. Basic barium petronate is a barium sulfonate salt of a 21-26 carbon atom hydrocarbon alkyl, and can be obtained, for example, from Chemtura. An example isopropyl amine sulphonate salt is dodecyl benzene sulfonic acid isopropyl
20 amine, which is available from Croda.

In some examples, the charge director constitutes about 0.001% to 20% by weight, in some examples 0.01% to 20% by weight, in some examples 0.01% to 10% by weight, in some examples 0.01% to 5% by weight of the total solids of a liquid electrostatic ink composition. In some examples, the charge director constitutes about 1% to 4% by
25 weight of the total solids of the liquid electrostatic ink composition, in some examples 2% to 4% by weight of the total solids of the electrostatic ink composition.

In some examples, the charge director is present in an amount sufficient to achieve a particle conductivity of 500 pmho/cm or less, in some examples, 450 pmho/cm or less, in some examples, 400 pmho/cm or less, in some examples, 350 pmho/cm or less, in
30 some examples, 300 pmho/cm or less, in some examples, 250 pmho/cm or less, in some examples, 200 pmho/cm or less, in some examples, 190 pmho/cm or less, in some examples, 180 pmho/cm or less, in some examples, 170 pmho/cm or less, in some examples, 160 pmho/cm or less, in some examples, 150 pmho/cm or less, in

some examples, 140 pmho/cm or less, in some examples, 130 pmho/cm or less, in some examples, 120 pmho/cm or less, in some examples, 110 pmho/cm or less, in some examples, about 100 pmho/cm. In some examples, the charge director is present in an amount sufficient to achieve a particle conductivity of 50 pmho/cm or more, in some examples, 60 pmho/cm or more, in some examples, 70 pmho/cm or more, in some examples, 80 pmho/cm or more, in some examples, 90 pmho/cm or more, in some examples, about 100 pmho/cm, in some examples, 150 pmho/cm or more, in some examples, 200 pmho/cm or more, in some examples, 250 pmho/cm or more, in some examples, 300 pmho/cm or more, in some examples, 350 pmho/cm or more, in some examples, 400 pmho/cm or more, in some examples, 450 pmho/cm or more, in some examples, 500 pmho/cm or more. In some examples, the charge director is present in an amount sufficient to achieve a particle conductivity of 50 pmho/cm to 500 pmho/cm, in some examples, 60 pmho/cm to 450 pmho/cm, in some examples, 70 pmho/cm to 400 pmho/cm, in some examples, 80 pmho/cm to 350 pmho/cm, in some examples, 90 pmho/cm to 300 pmho/cm, in some examples, 100 pmho/cm to 250 pmho/cm, in some examples, 110 pmho/cm to 200 pmho/cm, in some examples, 120 pmho/cm to 500 pmho/cm, in some examples, 130 pmho/cm to 450 pmho/cm, in some examples, 140 pmho/cm to 400 pmho/cm, in some examples, 150 pmho/cm to 350 pmho/cm, in some examples, 160 pmho/cm to 300 pmho/cm.

20 In some examples, the charge director is present in an amount of from 3 mg/g to 50 mg/g, in some examples from 3 mg/g to 45 mg/g, in some examples from 10 mg/g to 40 mg/g, in some examples from 5 mg/g to 35 mg/g, in some examples, 20 mg/g to 35 mg/g, in some examples, 22 mg/g to 34 mg/g (where mg/g indicates mg per gram of solids of the liquid electrostatic ink composition).

25 *Charge adjuvant*

In some examples, the LEP ink composition further includes a charge adjuvant. A charge adjuvant may promote charging of the particles when a charge director is present. The method as described herein may involve adding a charge adjuvant at any stage. The charge adjuvant can include, for example, barium petronate, calcium petronate, Co salts of naphthenic acid, Ca salts of naphthenic acid, Cu salts of naphthenic acid, Mn salts of naphthenic acid, Ni salts of naphthenic acid, Zn salts of naphthenic acid, Fe salts of naphthenic acid, Ba salts of stearic acid, Co salts of stearic acid, Pb salts of stearic acid, Zn salts of stearic acid, Al salts of stearic acid, Zn salts of stearic acid, Cu salts of stearic acid, Pb salts of stearic acid, Fe salts of stearic acid,

metal carboxylates (e.g., Al tristearate, Al octanoate, Li heptanoate, Fe stearate, Fe distearate, Ba stearate, Cr stearate, Mg octanoate, Ca stearate, Fe naphthenate, Zn naphthenate, Mn heptanoate, Zn heptanoate, Ba octanoate, Al octanoate, Co octanoate, Mn octanoate, and Zn octanoate), Co lineolates, Mn lineolates, Pb lineolates, Zn lineolates, Ca oleates, Co oleates, Zn palmirate, Ca resinates, Co resinates, Mn resinates, Pb resinates, Zn resinates, AB diblock copolymers of 2-ethylhexyl methacrylate-co- methacrylic acid calcium and ammonium salts, copolymers of an alkyl acrylamidoglycolate alkyl ether (e.g., methyl acrylamidoglycolate methyl ether- co-vinyl acetate), or hydroxy bis(3,5-di-tert-butyl salicylic) aluminate monohydrate. In an example, the charge adjuvant is or includes aluminum di- or tristearate. In some examples, the charge adjuvant is VCA (aluminium stearate and aluminium palmitate, available from Sigma Aldrich).

The charge adjuvant may be present in an amount of about 0.1% to 5% by weight, in some examples about 0.1% to 1% by weight, in some examples about 0.3% to 0.8% by weight of the total solids of the liquid electrostatic ink composition, in some examples, about 1 wt.% to 5 wt.% of the total solids of the liquid electrostatic ink, in some examples about 1 wt.% to 3 wt.% of the total solids of the liquid electrostatic ink composition, in some examples about 1.5 wt.% to 2.5 wt.% of the total solids of the liquid electrostatic ink composition.

The charge adjuvant may be present in an amount of less than 5% by weight of total solids of the liquid electrostatic ink composition, in some examples in an amount of less than 4.5% by weight, in some examples in an amount of less than 4% by weight, in some examples in an amount of less than 3.5% by weight, in some examples in an amount of less than 3% by weight, in some examples in an amount of less than 2.5% by weight, in some examples, in an amount of less than 2% by weight of the total solids of the liquid electrostatic ink composition.

In some examples, the liquid electrostatic ink composition further includes, e.g. as a charge adjuvant, a salt of multivalent cation and a fatty acid anion. The salt of multivalent cation and a fatty acid anion can act as a charge adjuvant. The multivalent cation may, in some examples, be a divalent or a trivalent cation. In some examples, the multivalent cation is selected from Group 2, transition metals and Group 3 and Group 4 in the Periodic Table. In some examples, the multivalent cation includes a metal selected from Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Al and Pb. In some examples, the multivalent cation is Al^{3+} . The fatty acid anion may be selected from a

saturated or unsaturated fatty acid anion. The fatty acid anion may be selected from a C8 to C26 fatty acid anion, in some examples a C14 to C22 fatty acid anion, in some examples a C16 to C20 fatty acid anion, in some examples a C17, C18 or C19 fatty acid anion. In some examples, the fatty acid anion is selected from a caprylic acid anion, capric acid anion, lauric acid anion, myristic acid anion, palmitic acid anion, stearic acid anion, arachidic acid anion, behenic acid anion and cerotic acid anion.

The charge adjuvant, which may, for example, be or include a salt of a multivalent cation and a fatty acid anion, may be present in an amount of 0.1 wt.% to 5 wt.% of the total solids of the liquid electrostatic ink composition, in some examples in an amount of 0.1 wt.% to 3 wt.% of the total solids of the liquid electrostatic ink composition, in some examples about 1 wt.% to 3 wt.% of the total solids of the liquid electrostatic ink composition, in some examples about 1.5 wt.% to 2.5 wt.% of the total solids of the liquid electrostatic ink composition.

Additives

The liquid electrophotographic (LEP) ink composition may include another additive or a plurality of other additives. The other additive or plurality of other additives may be added at any stage of the method. The other additive or plurality of other additives may be selected from a charge adjuvant, a wax, a surfactant, viscosity modifiers, and compatibility additives. The wax may be an incompatible wax. As used herein, "incompatible wax" may refer to a wax that is incompatible with the resin. Specifically, the wax phase separates from the resin phase upon the cooling of the resin fused mixture on a print substrate during and after the transfer of the ink film to the print substrate, e.g. from an intermediate transfer member, which may be a heated blanket. In some examples, the LEP ink composition comprises silica, which may be added, for example, to improve the durability of images produced using the LEP ink. The other additives may constitute 10 wt.% or less of the total solids of the electrostatic ink composition, in some examples, 5 wt.% or less of the total solids of the electrostatic ink composition, in some examples, 3 wt.% or less of the total solids of the electrostatic ink composition.

30 Method of producing a material with a perovskite structure on a substrate

In an aspect, there is provided a method of producing a material with a perovskite structure on a substrate. In some examples, the material with a perovskite structure has a chemical formula selected from ABX_3 and A_2BX_6 . In some examples, the material

with a perovskite structure has the chemical formula ABX_3 . In some examples, the material with a perovskite structure has the chemical formula A_2BX_6 . In some examples, there is provided a method of producing a material with a perovskite structure on a substrate by liquid electrophotographic (LEP) printing.

- 5 In some examples, the method of producing a material with a perovskite structure on a substrate by liquid electrophotographic (LEP) printing may comprise either liquid electrophotographically printing onto the substrate a composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid; liquid electrophotographically printing onto the substrate a composition comprising a
- 10 dispersion of a salt selected from BX_2 and BX_4 and a thermoplastic resin in a carrier liquid; and heat treating the printed compositions to form the material with a perovskite structure; or liquid electrophotographically printing onto the substrate a composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid to form the material with a perovskite structure.
- 15 In some examples, A is a cation, B is a cation and X is an anion. In some examples, the material with a perovskite structure has the chemical formula ABX_3 , wherein A is a monovalent cation, B is a divalent cation and X is a monovalent anion. In some examples, the material with a perovskite structure has the chemical formula ABX_3 , wherein A is a divalent cation, B is a tetravalent cation and X is a monovalent cation. In
- 20 some examples, the material with a perovskite structure has the chemical formula A_2BX_6 , wherein A is a monovalent cation, B is a tetravalent cation and X is a monovalent anion.

In some examples, the thermoplastic resin comprises a copolymer of an alkylene monomer and a monomer having acidic side groups.

- 25 In some examples, the method of producing a material with a perovskite structure on a substrate by liquid electrophotographic (LEP) printing may comprise liquid electrophotographically printing onto the substrate a composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid; liquid electrophotographically printing onto the substrate a composition comprising a
- 30 dispersion of a salt selected from BX_2 and BX_4 and a thermoplastic resin in a carrier liquid; and heat treating the printed compositions to form the material with a perovskite structure. In some examples, the method of producing a material with a perovskite structure on a substrate by liquid electrophotographic (LEP) printing may comprise

liquid electrophotographically printing onto the substrate a composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid; liquid electrophotographically printing onto the substrate a composition comprising a dispersion of a salt BX₂ and a thermoplastic resin in a carrier liquid; and heat treating
5 the printed compositions to form the material with a perovskite structure. In some examples, the method of producing a material with a perovskite structure on a substrate by liquid electrophotographic (LEP) printing may comprise liquid electrophotographically printing onto the substrate a composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid; liquid
10 electrophotographically printing onto the substrate a composition comprising a dispersion of a salt BX₄ and a thermoplastic resin in a carrier liquid; and heat treating the printed compositions to form the material with a perovskite structure.

In some examples, the heat treatment may comprise heating the printed compositions at 50°C or more, for example, 60°C or more, 70°C or more, 80°C or more, 90°C or
15 more, 100°C or more, 110°C or more, 120°C or more, 130°C or more, 140°C or more, 150°C or more, 160°C or more, 170°C or more, 180°C or more, 190°C or more, 200°C or more. In some examples, the heat treatment may comprise heating the printed compositions at 200°C or less, for example, 190°C or less, 180°C or less, 170°C or less, 160°C or less, 150°C or less, 140°C or less, 130°C or less, 120°C or less, 110°C
20 or less, 100°C or less, 90°C or less, 80°C or less, 70°C or less, 60°C or less, or 50°C or less. In some examples, the heat treatment may comprise heating the printed compositions at 50°C to 200°C, for example, 60°C to 190°C, 70°C to 180°C, 80°C to 170°C, 90°C to 160°C, 90°C to 150°C, 100°C to 140°C, 50°C to 130°C, 70°C to 120°C, 80°C to 110°C or 90°C to 100°C. In some examples, the heat treatment may comprise
25 heating for 30 s or more, for example, 40 s or more, 50 s or more, 1 min or more, 1 min 10 s or more, 1 min 20 s or more, 1 min 30 s or more, 1 min 40 s or more, 1 min 50 s or more, 2 min or more, 2 min 10 s or more, 2 min 20 s or more, 2 min 30 s or more, 2 min 40 s or more, 2 min 50 s or more, or 3 min or more. In some examples, the heat treatment may comprise heating for 3 min or less, for example, 2 min 50 s or less, 2
30 min 40 s or less, 2 min 30 s or less, 2 min 20 s or less, 2 min 10 s or less, 2 min or less, 1 min 50 s or less, 1 min 40 s or less, 1 min 30 s or less, 1 min 20 s or less, 1 min 10 s or less, 1 min or less, 50 s or less, 40 s or less, or 30 s or less. In some examples, the heat treatment may comprise heating for 30 s to 3 min, for example, 40 s to 2 min 50 s, 50 s to 2 min 40 s, 1 min to 2 min 30 s, 1 min 10 s to 2 min 20 s, 1 min 20 s to 2

min 10 s, 1 min 30 s to 2 min, 1 min 40 s to 3 min, 1 min 50 s to 2 min 30 s, 1 min 50 s to 2 min 20 s, or 2 min to 2 min 10 s.

In some examples, the method of producing a material with a perovskite structure on a substrate by liquid electrophotographic (LEP) printing may comprise liquid
5 electrophotographically printing onto the substrate a composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid to form the material with a perovskite structure.

In some examples, the method of producing a material with a perovskite structure on a substrate by liquid electrophotographic (LEP) printing may comprise liquid
10 electrophotographically printing onto the substrate a composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid to form the material with a perovskite structure, wherein the composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid was formed by combining a liquid electrophotographic ink
15 composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid with a liquid electrophotographic ink composition comprising a dispersion of a salt selected from BX_2 and BX_4 and a thermoplastic resin in a carrier liquid; or by dispersing a salt AX, a salt selected from BX_2 and BX_4 , and a thermoplastic resin in a carrier liquid.

20 In some examples, the method of producing a material with a perovskite structure on a substrate by liquid electrophotographic (LEP) printing may comprise liquid electrophotographically printing onto the substrate a composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid to form the material with a perovskite structure, wherein the composition
25 comprising a dispersion of the material with a perovskite structure and a thermoplastic resin in a carrier liquid was formed by combining a liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid with a liquid electrophotographic ink composition comprising a dispersion of a salt selected from BX_2 and BX_4 and a thermoplastic resin in a carrier liquid.

30 In some examples, the method of producing a material with a perovskite structure on a substrate by liquid electrophotographic (LEP) printing may comprise liquid electrophotographically printing onto the substrate a composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a

carrier liquid to form the material with a perovskite structure, wherein the composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid was formed by dispersing a salt AX, a salt selected from BX₂ and BX₄, and a thermoplastic resin in a carrier liquid.

- 5 In some examples, the material with a perovskite structure on the substrate may have a thickness of 0.1 μm or more, for example, 0.2 μm or more, 0.3 μm or more, 0.4 μm or more, 0.5 μm or more, 0.6 μm or more, 0.7 μm or more, 0.8 μm or more, 0.9 μm or more, 1 μm or more. In some examples, the material with a perovskite structure on the substrate may have a thickness of 1 μm or less, for example, 0.9 μm or less, 0.8 μm or less, 0.7 μm or less, 0.6 μm or less, 0.5 μm or less, 0.4 μm or less, 0.3 μm or less, 0.2 μm or less, 0.1 μm or less. In some examples, the material with a perovskite structure on the substrate may have a thickness of 0.1 μm to 1 μm for example, 0.2 μm to 0.9 μm, 0.3 μm to 0.8 μm, 0.4 μm to 0.7 μm, or 0.5 μm to 0.6 μm.

- 15 In some examples, the material with a perovskite structure on the substrate may form the light harvesting active layer of a photovoltaic cell.

In some examples, liquid electrophotographically printing a composition onto the substrate comprises contacting the composition with a latent electrostatic image on a surface to create a developed image and transferring the developed image to the substrate, in some examples, via an intermediate transfer member.

- 20 In some examples, the surface on which the (latent) electrostatic image is formed or developed may be a rotating member, for example, in the form of a cylinder. The surface on which the (latent) electrostatic image is formed or developed may form a part of a photo imaging plate (PIP). The method may involve passing the ink composition between a stationary electrode and a rotating member, which may be a member having the surface having the (latent) electrostatic image thereon or a member in contact with the surface having the (latent) electrostatic image thereon. A voltage is applied between the stationary electrode and the rotating member, such that particles adhere to the surface of the rotating member. The intermediate transfer member, if present, may be a rotating flexible member, which may be heated, for example, to a temperature of from 80 to 160°C.
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- 30

Method of producing the liquid electrophotographic ink composition comprising AX

In some examples, the method of producing a liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid comprises combining the salt AX, the thermoplastic resin and the carrier liquid. In
5 some examples, the method of producing the liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid comprises dispersing a salt AX and a thermoplastic resin in a carrier liquid.

In some examples, the method of producing a liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier
10 liquid may comprise combining the thermoplastic resin with the carrier liquid before adding the salt AX to the combined thermoplastic resin and carrier liquid. In some examples, the method of producing a liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid may comprise dispersing the thermoplastic resin in the carrier liquid before adding the salt
15 AX to the dispersion of the thermoplastic resin in the carrier liquid.

In some examples, combining the salt AX with the thermoplastic resin and the carrier liquid may comprise grinding the salt AX and the thermoplastic resin in the presence of the carrier liquid.

In some examples, the method of producing a liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier
20 liquid may comprise adding a charge adjuvant to the liquid electrophotographic ink composition. In some examples, the charge adjuvant may be added to the liquid electrophotographic ink composition before, during or after the salt AX, the thermoplastic resin and the carrier liquid are combined. In some examples, the charge
25 adjuvant may be added to the liquid electrophotographic ink composition before, during or after addition of the salt AX to the combined thermoplastic resin and carrier liquid.

In some examples, the method of producing a liquid electrophotographic (LEP) ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid may comprise adding a charge director to the liquid electrophotographic ink
30 composition. In some examples, the charge director may be added to the liquid electrophotographic ink composition before, during or after the salt AX, the thermoplastic resin and the carrier liquid are combined. In some examples, the charge director may be added to the liquid electrophotographic ink composition before, during

or after addition of the salt AX to the combined thermoplastic resin and carrier liquid. In some examples, the charge director may be added to the liquid electrophotographic ink composition before, during or after the charge adjuvant is added to the liquid electrophotographic ink composition.

- 5 In some examples, the method of producing an LEP ink composition may comprise suspending a thermoplastic resin in a carrier liquid before adding the salt AX. In some examples, the method may comprise suspending a first thermoplastic resin and a second thermoplastic resin in a carrier liquid. In some examples, the LEP ink composition comprises chargeable particles comprising a first thermoplastic resin and
10 a second thermoplastic resin. In some examples, the method of producing an LEP ink composition may comprise suspending chargeable particles comprising a first thermoplastic resin and a second thermoplastic resin in a carrier liquid.

- In some examples, the method of producing an LEP ink composition may comprise dispersing a first thermoplastic resin and a second thermoplastic resin in a carrier
15 liquid. In some examples, the method of producing an LEP ink composition may comprise dispersing chargeable particles comprising a first thermoplastic resin and a second thermoplastic resin in a carrier liquid.

- In some examples, the method of producing an LEP ink composition may comprise combining a thermoplastic resin (for example, the first thermoplastic resin) with the
20 carrier liquid and subsequently adding the other resin (for example, the second resin). In some examples, the method of producing an LEP ink composition comprises combining a resin (for example, the first resin) with the carrier liquid to form a paste and subsequently adding the other resin (for example, the second resin). In some examples, the resin and the carrier liquid are combined and heated to an elevated
25 temperature before adding the other resin, which may have also been heated to an elevated temperature. In some examples, the resin and the carrier liquid are combined and heated to a temperature above the melting point of the resin before adding the other resin, which may have also been heated to a temperature above its melting point. In some examples, the resin and carrier liquid are combined and heated until the resin
30 has melted and/or dissolved in the carrier liquid before adding the other resin. In some examples, adding the other resin to the combined resin and carrier liquid comprises mixing the other resin with the combined resin and carrier liquid.

The melting point of the resin may be determined by differential scanning calorimetry, for example, using ASTM D3418.

In some examples, the resin and the carrier liquid are combined and heated to a temperature of at least 70°C, for example, at least 80°C, for example, at least 90°C, for example, at least 100°C, for example, at least 110°C, for example, at least 120°C, for example, 130°C, for example, to melt the resin. In some examples, the other resin is heated before being added to the combined resin and carrier liquid. In some examples, the other resin is heated to at least 30°C, in some examples, at least 40°C, in some examples, at least 45°C, in some examples, at least 50°C before being added to the combined resin and carrier liquid. In some examples, the other resin is heated to 100°C or less, in some examples, 90°C or less, in some examples, 80°C or less, in some examples, 75°C or less, in some examples, 70°C or less, in some examples, 60°C or less before being added to the combined resin and carrier liquid. In some examples, the other resin is heated to reduce the viscosity of the other resin before being added to the first resin and the carrier liquid.

In some examples, the method comprises combining the first resin with the carrier liquid to form a first composition; combining the second resin with the carrier liquid to form a second composition; and subsequently combining the first composition and the second composition to form a liquid electrophotographic ink composition. In some examples, the method comprises combining the first resin with the carrier liquid to form a first paste; combining the second resin with the carrier liquid to form a second paste; and subsequently combining the first paste and the second paste to form a liquid electrophotographic ink composition. In some examples, the first resin and the carrier liquid are combined and heated to an elevated temperature to form a first heated composition; the second resin and the carrier liquid are combined and heated to an elevated temperature to form a second heated composition; and subsequently the first heated composition and the second heated composition are combined. In some examples, the first resin and the carrier liquid are combined and heated to a temperature above the melting point of the first resin to form a first heated composition; the second resin and the carrier liquid are combined and heated to a temperature above the melting point of the second resin to form a second heated composition; and subsequently the first heated composition and the second heated composition are combined. In some examples, the first composition and the second composition are heated to the same temperature, which may be a temperature above the melting temperature of all of the resins.

In some examples, the method of producing an LEP ink composition comprises mixing the first resin and the second resin together and then combining the mixture of the resins with the carrier liquid.

5 In some examples, the first resin and the second resin are combined with the carrier liquid and subsequently heated to an elevated temperature. In some examples, the first resin and the second resin are combined with the carrier liquid and subsequently heated to a temperature above the melting point of at least one, optionally all, of the resins. In some examples, the first resin and the second resin are combined with the carrier liquid and subsequently heated to a temperature of at least 70°C, for example,
10 at least 80°C, for example, at least 90°C, for example, at least 100°C, for example, at least 110°C, for example, at least 120°C, for example, 130°C, for example, to melt at least one, optionally all, of the resins. In some examples, the combined first resin, second resin and carrier liquid are heated until all of the resins have melted and/or dissolved in the carrier liquid.

15 In some examples, the method of producing a liquid electrophotographic ink composition comprises combining a first resin, a second resin, and a carrier liquid.

In some examples, the chargeable particles comprise the first resin and the second resin.

20 Melting and/or dissolving a resin (or resins) in the carrier liquid may result in the carrier fluid appearing clear and homogeneous. In some examples, the resin (or resins) and carrier liquid are heated before, during or after mixing.

In some examples, the resin (or resins) and the carrier liquid are mixed at a mixing rate of 500 rpm or less, for example, 400 rpm or less, for example, 300 rpm or less, for example, 200 rpm or less, for example, 100 rpm or less, for example, 75 rpm or less,
25 for example, 50 rpm. In some examples, mixing may continue until melting and/or dissolution of the resin (or resins) in the carrier liquid is complete.

In some examples, after combining and heating the resins and the carrier liquid, the mixture is cooled to a temperature below the melting point of the resins, for example, to room temperature. In some examples, the chargeable particles are removed from the carrier liquid and re-dispersed in a new portion of carrier liquid, which may be the same
30 or a different carrier liquid.

In some examples, the method of producing an LEP ink composition comprises adding a salt AX to the combined first resin, second resin and carrier liquid. In some examples, the method of producing an LEP ink composition comprises adding a salt AX to the combined first resin, second resin and carrier liquid to form chargeable particles comprising the resins and a salt AX. In some examples, the method of producing an LEP ink composition comprises grinding the salt AX and the resins in the presence of the carrier liquid to form a paste. In some examples, the method of producing an LEP ink composition comprises heating and mixing the salt AX and the resins in the presence of the carrier liquid to form a paste.

- 5
- 10 In some examples, the method of producing an LEP ink composition comprises adding a charge adjuvant to the combined first resin, second resin and carrier liquid and optionally grinding. In some examples, the method of producing an LEP ink composition comprises adding a charge adjuvant and a salt AX to the combined first resin, second resin and carrier liquid and optionally grinding. In some examples, the
- 15 method of producing an LEP ink composition comprises adding a charge adjuvant to the combined first resin, second resin, salt AX and carrier liquid and optionally grinding.

- In some examples, the method of producing an LEP ink composition comprises grinding at a grinding speed of at least 50 rpm. In some examples, the method of producing an LEP ink composition comprises grinding at a grinding speed of up to
- 20 about 600 rpm. In some examples, the method of producing an LEP ink composition comprises grinding for at least 1 h, in some examples, for at least 2 h. In some examples, the method of producing an LEP ink composition comprises grinding for up to about 12 h. In some examples, the method of producing an LEP ink composition comprises grinding at a temperature of at least about 30°C, for example, at least about
- 25 35°C, for example, at least about 40°C, for example, at least about 50°C. In some examples, the method of producing an LEP ink composition comprises grinding at a temperature of at least about 50°C for a first time period, in some examples, for at least 1 h, in some examples, for at least 1.5 h and then reducing the temperature to a temperature of at least 30°C, in some examples, at least 35°C and continuing grinding
- 30 for at least 5 h, in some examples, at least 9 h, in some examples, at least 10 h.

In some examples, the method of producing an LEP ink composition comprises adding a charge director to the combined first resin, second resin and carrier liquid. In some examples, the method of producing an LEP ink composition comprises adding a charge director to the combined first resin, second resin, salt AX and carrier liquid. In some

examples, the method of producing an LEP ink composition comprises adding a charge director to the combined first resin, second resin, charge adjuvant and carrier liquid. In some examples, the method of producing an LEP ink composition comprises adding a charge director to the combined first resin, second resin, salt AX, charge adjuvant and carrier liquid.

Method of producing the liquid electrophotographic ink composition comprising BX_2

In some examples, the liquid electrophotographic ink composition comprising a dispersion of a salt BX_2 and a thermoplastic resin in a carrier liquid may be produced by the method described above for producing the liquid electrophotographic ink composition comprising the salt AX except that the salt BX_2 is used instead of the salt AX.

Method of producing the liquid electrophotographic ink composition comprising BX_4

In some examples, the liquid electrophotographic ink composition comprising a dispersion of a salt BX_4 and a thermoplastic resin in a carrier liquid may be produced by the method described above for producing the liquid electrophotographic ink composition comprising the salt AX except that the salt BX_4 is used instead of the salt AX.

Method of producing the liquid electrophotographic ink composition comprising a material with a perovskite structure

In some examples, the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid may be produced by the method described above for producing the liquid electrophotographic ink composition comprising the salt AX except that a combination of the salt AX and a salt selected from BX_2 and BX_4 is used instead of the salt AX. In some examples, the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid (wherein the material with a perovskite structure has the chemical formula ABX_3) may be produced by the method described above for producing the liquid electrophotographic ink composition comprising the salt AX except that the salts AX and BX_2 are used in a 1:1 ratio (by number of moles of each salt) instead of the salt AX. In some examples, the liquid electrophotographic ink composition comprising a dispersion of material with a perovskite structure and a thermoplastic resin in a carrier

liquid (wherein the material with a perovskite structure has the chemical formula A_2BX_6) may be produced by the method described above for producing the liquid electrophotographic ink composition comprising the salt AX except that the salts AX and BX_4 are used in a 2:1 ratio (by number of moles of each salt) instead of the salt AX.

In some examples, the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid may be produced by combining a composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid with a composition comprising a dispersion of a salt selected from BX_2 and BX_4 and a thermoplastic resin in a carrier liquid. In some examples, the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid (wherein the material with a perovskite structure has the chemical formula ABX_3) may be produced by combining, in a 1:1 ratio (by number of moles of each salt), a composition (for example, a liquid electrophotographic ink composition) comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid with a composition (for example, a liquid electrophotographic ink composition) comprising a dispersion of a salt BX_2 and a thermoplastic resin in a carrier liquid. In some examples, the liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid (wherein the material with a perovskite structure has the chemical formula A_2BX_6) may be produced by combining, in a 2:1 ratio (by number of moles of each salt), a composition (for example, a liquid electrophotographic ink composition) comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid with a composition (for example, a liquid electrophotographic ink composition) comprising a dispersion of a salt BX_4 and a thermoplastic resin in a carrier liquid.

In some examples, the liquid electrophotographic ink composition comprising a dispersion of material with a perovskite structure and a thermoplastic resin in a carrier liquid may be produced by combining, in a stoichiometric ratio, a composition (for example, a liquid electrophotographic ink composition) comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid with a composition (for example, a liquid electrophotographic ink composition) comprising a dispersion of a salt selected from BX_2 and BX_4 and a thermoplastic resin in a carrier liquid.

In some examples, the composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid was mixed by high shear mixing prior to being combined with the dispersion of a salt selected from BX_2 and BX_4 and a thermoplastic resin in a carrier liquid. In some examples, the composition comprising a dispersion of a salt selected from BX_2 and BX_4 and a thermoplastic resin in a carrier liquid was mixed by high shear mixing prior to being combined with the dispersion of a salt AX and a thermoplastic resin in a carrier liquid. In some examples, the composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid and the composition comprising a dispersion of a salt selected from BX_2 and BX_4 and a thermoplastic resin in a carrier liquid were separately mixed by high shear mixing prior to the two compositions being combined. In some examples, high shear mixing comprises mixing at a shear rate of up to 35000 rpm, for example, up to 30000 rpm, or up to 25000 rpm. In some examples, high shear mixing comprises mixing at a shear rate of at least 7000 rpm, for example, at least 10000 rpm, at least 15000 rpm, at least 20000 rpm or at least 25000 rpm. In some examples, high shear mixing comprises mixing at a shear rate of 7000 rpm to 35000 rpm, for example, 10000 rpm to 30000 rpm, or 15000 rpm to 25000 rpm.

In some examples, combining a composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid with a composition comprising a dispersion of a salt BX_2 or a salt BX_4 and a thermoplastic resin in a carrier liquid may comprise mixing the two compositions for 10 min or less, for example, 5 min or less, 4 min or less, 3 min or less, 2 min or less, 1 min or less. In some examples, combining a composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid with a composition comprising a dispersion of a salt BX_2 or a salt BX_4 and a thermoplastic resin in a carrier liquid may comprise mixing the two compositions for 1 min or more, for example, 2 min or more, 3 min or more, 4 min or more, 5 min or more, 10 min or more. In some example, combining a composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid with a composition comprising a dispersion of a salt BX_2 and a thermoplastic resin in a carrier liquid may comprise mixing the two compositions for 1 min to 10 min, for example, 2 min to 5 min, 3 min to 4 min. In some examples, the mixing may be at room temperature, for example, about 25°C. In some examples, the mixing speed may be 50 rpm or less. In some examples, the mixing may be performed manually over a few minutes. In some examples, the mixture may then be rested for a period of time to form the material with a perovskite structure. In some examples, the period of time may be 1 h or more, for example, 1.5 h or more, 2 h or

more, 2.5 h or more, 3 h or more. In some examples, the period of time may be 3 h or less, for example, 2.5 h or less, 2 h or less, 1.5 h or less, or 1 h or less. In some examples, the period of time may be 1 h to 3 h, for example, 1.5 h to 2.5 h or 2 h to 2.5 h.

5 Printed substrate

In another aspect, there is provided a printed substrate. The printed substrate may comprise a substrate; and a liquid electrophotographically printed composition comprising a thermoplastic resin and a material with a perovskite structure disposed on the substrate. In some examples, the printed substrate may comprise a substrate; and
10 a liquid electrophotographically printed composition comprising a thermoplastic resin and a material with a perovskite structure disposed on the substrate; wherein the material with a perovskite structure has a chemical formula selected from ABX_3 and A_2BX_6 , wherein A is a cation, B is a cation and X is an anion; and wherein the thermoplastic resin comprises an alkylene monomer and a monomer having acidic side
15 groups.

In some examples, the liquid electrophotographically printed composition may form a layer having a thickness of 0.5 μm or more, for example, 0.6 μm or more, 0.7 μm or more, 0.8 μm or more, 0.9 μm or more, 1 μm or more. In some examples, the liquid electrophotographically printed composition may form a layer having a thickness of 1
20 μm or less, for example, 0.9 μm or less, 0.8 μm or less, 0.7 μm or less, 0.6 μm or less, 0.5 μm or less. In some examples, the liquid electrophotographically printed material may form a layer having a thickness of 0.5 μm to 1 μm , for example, 0.6 μm to 0.9 μm , 0.7 μm to 0.8 μm .

In some examples, the printed substrate may be a photovoltaic cell or an electronic
25 device. In some examples, the printed substrate may be a photovoltaic cell, wherein the liquid electrophotographically printed composition forms the light harvesting active layer of the photovoltaic cell. In some examples, the printed substrate may be an electronic device, wherein the liquid electrophotographically printed composition forms a semiconductor component of the electronic device.

30 In some examples, the printed substrate may comprise a conductive substrate, an electron transport layer disposed on the conductive substrate, a light harvesting active layer disposed on the electron transport layer; a hole transport layer disposed on the light harvesting active layer; and a conductive layer disposed on the hole transport

layer. In some examples, the material with a perovskite structure may be the light harvesting active layer.

Substrate

5 In some examples, the substrate may comprise any material capable of being liquid electrophotographically printed. In some examples, the substrate may comprise an electron transport layer for use in a photovoltaic cell.

10 In some examples, the substrate may comprise an electron transport layer. In some examples, the electron transport layer may be ZnO, alumina or titania. In some examples, the material with a perovskite structure may be disposed on the electron transport layer of the substrate.

In some examples, the electron transport layer may be disposed on the surface of an indium tin oxide coated polyethylene terephthalate layer.

In some examples, the substrate may be plastic, for example, polyethylene terephthalate.

15 EXAMPLES

The following illustrates examples of the methods and other aspects described herein. Thus, these Examples should not be considered as limitations of the present disclosure, but are merely in place to teach how to make examples of the present disclosure.

20 Materials

Salts

CsBr: available from Sigma-Aldrich

SnBr₂: available from Sigma-Aldrich

Resins

25 Nucrel® 699: a copolymer of ethylene and methacrylic acid, made with nominally 11 wt.% methacrylic acid (available from DuPont).

AC-5120: a copolymer of ethylene and acrylic acid with an acrylic acid content of 15 wt.% (available from Honeywell).

Carrier Liquid

Isopar L™: an isoparaffinic oil comprising a mixture of C11-C13 isoalkanes (produced
5 by Exxon Mobil™; CAS number 64742-48-9.

Charge Adjuvant

Aluminium stearate: available from Sigma-Aldrich

Charge Director

10 NCD (natural charge director): KT (natural soya lecithin in phospholipids and fatty acids), BBP (basic barium petronate, i.e., a barium sulfonate salt of a 21-26 carbon hydrocarbon alkyl, available from Cemtura™), and GT (dodecyl benzene sulfonic acid isopropyl amine, supplied by Croda™). The composition being 6.6 wt.% KT, 9.8 wt.% BBP and 3.6 wt.% GT and balance (80 wt.%) Isopar L™.

Resin Paste

15 Nucrel™ 699 and AC™-5120 were combined in a ratio of 4:1 with Isopar™ L (from Chevron) at 42 wt.% solids. The mixture was mixed at 50 rpm in a Ross mixer and heated to 120°C until a clear one phase solution (approximately 2 hours) was produced. The mixture was then cooled slowly to below 60°C to give the swollen resin in Isopar™ L as a paste. The paste was diluted with Isopar™ L to 25 wt.% NVS.

20 Liquid Electrophotographic Ink Composition 1 – CsBr

CsBr (2.5 g), the resin paste (9.2 g; at 25 wt.% NVS) and Al stearate (0.2 g) were combined with Isopar™ L in a glass container to give 100 g of a dispersion at 5 wt.% total NVS. To the glass container, 80 g of 0.9 mm ceramic media were added. The mixture was placed in a high powered shaker grinding tool (Fast&Fluid SK550 1.1
25 Shaker Skandex™) and was ground for 12 hours at 500 rpm to produce a CsBr-based liquid electrophotographic ink composition at 5 wt.% NVS. A charge director (NCD) in an amount of 50 mg/g of solids was added to provide the charged dispersion for liquid electrophotographic printing.

Liquid Electrophotographic Ink Composition 2 – SnBr₂

SnBr₂ (3 g), resin paste (7.2 g; at 25 wt.% NVS) and Al stearate (0.2 g) were combined with Isopar™ L in a glass container to give a 100 g dispersion at 5 wt.% total NVS. To the glass container, 80 g of 0.9 mm ceramic media were added. The mixture was
5 placed in a high power shaker grinding tool (Fast&Fluid SK550 1.1 Shaker Skandex™) and was ground for 12 hours at 500 rpm to produce a SnBr₂-based liquid electrophotographic ink composition at 5 wt.% NVS. A charge director (NCD) in an amount of 50 mg/g of solids was added to provide the charged dispersion for liquid electrophotographic printing.

10 Example 1 – in situ film formation of perovskite

Liquid electrophotographic ink composition 1 (a white colored ink comprising CsBr) was electroplated onto a polyethylene terephthalate (PET) substrate to form a layer with a thickness of typically 0.5 μm to 1 μm. Liquid electrophotographic ink composition 2 (comprising SnBr₂) was then applied by electroplating to form a layer with a thickness
15 of typically 0.5 μm to 1 μm. The two layers were fused and dried on a hot plate at 100°C for 2 min to give a colored perovskite film. It is believed that the heating provides the activation energy for the formation of the perovskite structure on the surface of the substrate.

Electroplating exploits the same phenomenon as liquid electrophotographic printing
20 and has therefore been used to demonstrate that these ink compositions are capable of being liquid electrophotographically printed. In electroplating, two electrodes are placed in the liquid electrophotographic ink composition and a strong electric field is applied between the two electrodes. The substrate is attached to the positively charged electrode. The chargeable particles of the liquid electrophotographic ink composition
25 are attracted to the positively charged electrode, forming a layer of the liquid electrophotographic ink composition on the substrate attached to the positively charged electrode. In liquid electrophotographic printing, a positively charged latent image is formed on the photoimaging plate (PIP) and the chargeable particles are attracted to the positively charged portions of the PIP. Thus, the formation of a layer of liquid
30 electrophotographic ink composition on the substrate during electroplating demonstrates that the composition is capable of being used as a liquid electrophotographic ink composition.

Liquid Electrophotographic Ink Composition 3 – Mix of Ink Compositions 1 and 2

Liquid electrophotographic ink composition 1 and liquid electrophotographic ink composition 2 were combined in a stoichiometric ratio, manually mixed for a few minutes and placed on a shelf for 2 hours. The white composition changed color, forming a dark colored CsSnBr_3 -containing liquid electrophotographic ink composition at 5 wt.% NVS. A charge director (NCD) in an amount of 50 mg/g of solids was added to provide the charged dispersion for liquid electrophotographic printing. It is believed that the separate grinding of each liquid electrophotographic ink composition and manual mixing of the combined composition provides the activation energy for the formation of the perovskite structure.

Example 2

Liquid electrophotographic ink composition 3 was deposited onto a PET substrate by electroplating.

Liquid Electrophotographic Ink composition 4 – in situ formation of CsSnBr_3

CsBr (1.25 g), SnBr_2 (1.5 g), resin paste (9.2 g; at 25 wt.% NVS) and Al stearate (0.2 g) were combined in a glass container. The dispersion was diluted with Isopar™ L to form a 100 g dispersion at 5 wt.% total NVS. To the glass container, 80 g of 0.9 mm ceramic media were added, and the mixture was ground in a high-power shaking tool (Fast&Fluid SK550 1.1 Shaker Skandex™) for 12 hours at 500 rpm to form a dark CsSnBr_3 perovskite based liquid electrophotographic ink composition. A charge director (NCD) in an amount of 50 mg/g of solids was added to provide the charged dispersion for liquid electrophotographic printing. It is believed that the grinding procedure provides the activation energy for the formation of the perovskite structure.

Example 3

Liquid electrophotographic ink composition 4 was deposited onto a PET substrate by electroplating.

Test results

A sample of the liquid electrophotographic ink composition 3 was dried. A comparison of the X-ray diffraction pattern obtained for the sample with those of CsSnBr_3 and

Cs_2SnBr_6 (spectra obtained from a library of experimental XRD patterns) shows that a mixture of these two perovskite structures is present.

The dark color of the obtained perovskite mixtures in Isopar L (liquid electrophotographic ink compositions 3 and 4) was found to be stable for several
5 months. In contrast, solutions for use in other methods of obtaining perovskite materials (e.g., by spray or spin coating techniques) cannot be stored. It is believed that the increased stability of the perovskite structure within the dispersion is due to the absence of polar molecules that, if present, interrupt the perovskite structure. Moreover, the hydrophobic nature of the carrier liquid (Isopar L) and thermoplastic
10 resin in the composition prevent, to some extent, exposure of the perovskite structure to water.

Liquid electrophotographic ink compositions 1 (comprising CsBr) and 2 (comprising SnBr_2) were also found to be stable for several months.

While the invention has been described with reference to certain examples, those
15 skilled in the art will appreciate that various modifications, changes, omissions, and substitutions can be made without departing from the spirit of the disclosure. It is intended, therefore, that the invention be limited by the scope of the following claims. Unless otherwise stated, the features of any dependent claim can be combined with the features of any of the other dependent claims and any of the independent claims.

Claims

1. A method of producing a material with a perovskite structure on a substrate by liquid electrophotographic (LEP) printing, the method comprising:
- 5 either
- liquid electrophotographically printing onto the substrate a composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid;
- 10 liquid electrophotographically printing onto the substrate a composition comprising a dispersion of a salt selected from BX_2 and BX_4 and a thermoplastic resin in a carrier liquid; and
- heat treating the printed compositions to form the material with a perovskite structure;
- or
- 15 liquid electrophotographically printing onto the substrate a composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid to form the material with a perovskite structure;
- 20 wherein the material with a perovskite structure has a chemical formula selected from ABX_3 and A_2BX_6 ;
- wherein A is a cation, B is a cation and X is an anion;
- wherein the thermoplastic resin comprises a copolymer of an alkylene monomer and a monomer having acidic side groups.
2. The method according to claim 1, wherein A is selected from a monovalent metal cation, a monovalent organic cation, or a mixture thereof.
- 25 3. The method according to claim 1, wherein the salt selected from BX_2 and BX_4 is BX_2 and B is a divalent metal cation or wherein the salt selected from BX_2 and BX_4 is BX_4 and B is a tetravalent metal cation.
4. The method according to claim 1, wherein X is a halide ion, for example, selected from iodide, bromide, chloride and mixtures thereof.
- 30 5. The method according to claim 1, wherein A is selected from methylammonium (MA), formamidinium (FA), rubidium (Rb), caesium (Cs), and mixtures thereof.

6. The method according to claim 1, wherein B is selected from lead (Pb), germanium (Ge), tin (Sn), antimony (Sb), bismuth (Bi), copper (Cu), manganese (Mn), cobalt (Co) and mixtures thereof.
7. The method according to claim 1, wherein the composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid was formed by
combining a liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid with a liquid electrophotographic ink composition comprising a dispersion of a salt selected from BX_2 and BX_4 and a thermoplastic resin in a carrier liquid;
or
dispersing a salt AX, a salt selected from BX_2 and BX_4 , and a thermoplastic resin in a carrier liquid.
8. The method according to claim 1, wherein the heat treatment was performed at a temperature of 200°C or less.
9. The method according to claim 1, wherein the thermoplastic resin comprises a copolymer of ethylene and a monomer selected from acrylic acid and methacrylic acid.
10. The method according to claim 1, wherein the substrate comprises an electron transport layer onto which the material with a perovskite structure is applied.
11. The method according to claim 1, wherein the carrier liquid is a hydrocarbon.
12. The method according to claim 1, wherein the carrier liquid is a branched chain hydrocarbon comprising 5 to 15 carbon atoms.
13. An ink set comprising:
a liquid electrophotographic ink composition comprising a dispersion of a material with a perovskite structure and a thermoplastic resin in a carrier liquid;
or
a liquid electrophotographic ink composition comprising a dispersion of a salt AX and a thermoplastic resin in a carrier liquid; and
a liquid electrophotographic ink composition comprising a dispersion of a salt selected from BX_2 and BX_4 and a thermoplastic resin in a carrier liquid;

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wherein the material with a perovskite structure has a chemical formula selected from ABX_3 and A_2BX_6 ;

wherein A is a cation, B is a cation and X is an anion;

5 wherein the thermoplastic resin comprises a copolymer of an alkylene monomer and a monomer having acidic side groups.

14. The ink set according to claim 13, wherein the carrier liquid comprises a hydrocarbon.

15. A printed substrate comprising:

a substrate; and

10 a liquid electrophotographically printed composition comprising a thermoplastic resin and a material with a perovskite structure disposed on the substrate;

wherein the material with a perovskite structure has a chemical formula selected from ABX_3 and A_2BX_6 ;

wherein A is a cation, B is a cation and X is an anion; and

15 wherein the thermoplastic resin comprises an alkylene monomer and a monomer having acidic side groups.

INTERNATIONAL SEARCH REPORT		International application No. PCT/US 2019/061716														
A. CLASSIFICATION OF SUBJECT MATTER <i>C09D 11/10 (2014.01)</i> <i>G03G 17/00 (2006.01)</i> <i>G03G 17/02 (2006.01)</i> According to International Patent Classification (IPC) or to both national classification and IPC																
B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) C09D 11/02, 11/10, 11/03, G03G 16/00, G03G 7/00, 17/00-17/04 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) PatSearch (RUPTO internal), USPTO, PAJ, Esp@cenet, DWPI, EAPATIS, PATENTSCOPE																
C. DOCUMENTS CONSIDERED TO BE RELEVANT																
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.														
X	WO 2017/148538 A1 (HP INDIGO B.V.) 08.09.2017, claims, [0024], [0032], [0037]-[0039], [0057]-[0065], [0080], [0081]	1-15														
A	WO 2012/082129 A1 (BAR-HAIM GIL) 21.06.2012	1-15														
A	US 8440381 B2 (HEWLETT PACKARD DEVELOPMENT CO LP) 14.05.2013	1-15														
☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.																
<table><tr><td>* Special categories of cited documents:</td><td>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</td></tr><tr><td>"A" document defining the general state of the art which is not considered to be of particular relevance</td><td>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone</td></tr><tr><td>"D" document cited by the applicant in the international application</td><td>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art</td></tr><tr><td>"E" earlier document but published on or after the international filing date</td><td>"&" document member of the same patent family</td></tr><tr><td>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</td><td></td></tr><tr><td>"O" document referring to an oral disclosure, use, exhibition or other means</td><td></td></tr><tr><td>"P" document published prior to the international filing date but later than the priority date claimed</td><td></td></tr></table>			* Special categories of cited documents:	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention	"A" document defining the general state of the art which is not considered to be of particular relevance	"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone	"D" document cited by the applicant in the international application	"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art	"E" earlier document but published on or after the international filing date	"&" document member of the same patent family	"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)		"O" document referring to an oral disclosure, use, exhibition or other means		"P" document published prior to the international filing date but later than the priority date claimed	
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Date of the actual completion of the international search 31 July 2020 (31.07.2020)		Date of mailing of the international search report 06 August 2020 (06.08.2020)														
Name and mailing address of the ISA/RU: Federal Institute of Industrial Property, Berezhkovskaya nab., 30-1, Moscow, G-59, GSP-3, Russia, 125993 Facsimile No: (8-495) 531-63-18, (8-499) 243-33-37		Authorized officer I. Eliseeva Telephone No. 8 499 240 25 91														