Claims

1. An electroconductive paste comprising:

50–90 wt. % of a copper component comprising at least two types of copper particles each having different median particle diameters d_{50};

0.5–10 wt. % of a glass frit;

0.1–5% wt. % of adhesion promoter, which is at least one selected member from the group consisting of cuprous oxide, titanium oxide, zirconium oxide, boron resinate, zirconium resinate, amorphous boron, lithium phosphate, bismuth oxide, aluminum oxide, and zinc oxide; and

5–20 wt. % of an organic vehicle.

2. ~~An~~ The electroconductive paste according to claim 1, comprising:

50–90 wt. % of a copper particle;

0.5–1.5 wt. % of a glass frit;

0.1–5% wt. % of adhesion promoter, which is at least one selected member from the group consisting of cuprous oxide, titanium oxide, zirconium oxide, boron resinate, zirconium resinate, amorphous boron, lithium phosphate, bismuth oxide, aluminum oxide, and zinc oxide; and

5–20 wt. % of an organic vehicle.

3. The electroconductive paste according to claim 1 or 2, wherein the copper particle comprises at least one of:

a first copper particle having a median particle diameter (d_{50}) of ~~about~~ 2.0–2.9 μm, preferably ~~about~~ 2.5 μm;

a second copper particle having a median particle diameter (d_{50}) of ~~about~~ 3.0–3.9 μm, preferably ~~about~~ 3.5 μm; and

a third copper particle having a median particle diameter (d_{50}) of ~~about~~ 4.0–4.9 μm, preferably ~~about~~ 4.5 μm.

4. The electroconductive paste according to any one of the preceding claims, wherein the first copper particle has a specific surface area of ~~about~~ 0.25–0.5 m^2/g.

5. The electroconductive paste according to any of the preceding claims, wherein the second copper particle has a specific surface area of ~~about~~ 0.25–0.75 m^2/g.

6. The electroconductive paste according to any of the preceding claims, wherein the third copper particle has a specific surface area of ~~about~~ 0.25–0.50 m^2/g.

7. The electroconductive paste according to any of the preceding claims, wherein the first copper particle is ~~about~~ 45 to ~~about~~ 65 wt. % of the electroconductive paste.

8. The electroconductive paste according to any of the preceding claims, wherein the second copper particle is ~~about~~ 15 to ~~about~~ 30 wt. % of the electroconductive paste.

9. The electroconductive paste according to any of the preceding claims, wherein the third copper particle is ~~about~~ 5 to ~~about~~ 15 wt. % of the electroconductive paste.

10. The electroconductive paste according to any of the preceding claims, wherein the glass frit comprises ~~about~~ 45–65% Bi_2O_3, 25–45% SiO_2, and 1–15% B_2O_3, preferably 50–60% Bi_2O_3, 30–40% SiO_2, and 1–10% B_2O_3, most preferably 55–60% Bi_2O_3, 35–40% SiO_2, and 5–10% B_2O_3.

11. The electroconductive paste according to any of claims 1 or 3-10, wherein the glass frit is ~~about~~ 0.5 to ~~about~~ 5 wt. % of the electroconductive paste, more preferably ~~about~~ 0.5 to ~~about~~ 1.5 wt. % of the electroconductive paste.

12. The electroconductive paste according to any of the preceding claims, wherein the adhesion promoter is bismuth oxide.

13. The electroconductive paste according to any of the preceding claims, further comprising ~~about~~ 0.5 to ~~about~~ 3 wt. % copper oxide.

14. An article comprising:

(a) an aluminum nitride substrate; and

(b) an electroconductive electrode comprising an electroconductive paste according to any of the preceding claims.

15. A method of forming an electroconductive circuit comprising:

(i) depositing a first layer of base layer electroconductive paste according to any of claims 1-13 on an aluminum nitride substrate;

(ii) drying the aluminum nitride substrate with the deposited base layer electroconductive paste at a temperature at ~~about~~ 100°C to ~~about~~ 125°C for ~~about~~ 5 to ~~about~~ 10 minutes;

(iii) subjecting the deposited base layer electroconductive paste and the aluminum nitride substrate to a temperature of ~~about~~ 900°C to ~~about~~ 1000°C in a nitrogen atmosphere comprising from ~~about~~ 1 to ~~about~~ 20 ppm oxygen;

(iv) depositing a second layer of a top layer electroconductive paste according to any of claims 1-13 on the substrate;

(v) drying the aluminum nitride substrate with the deposited top layer electroconductive paste at a temperature at ~~about~~ 100°C to ~~about~~ 125°C for ~~about~~ 5 to ~~about~~ 10 minutes; and

(vi) subjecting the deposited layers and the aluminum nitride substrate to a temperature of ~~about~~ 900°C to ~~about~~ 1000°C in a nitrogen atmosphere comprising from ~~about~~ 1 to ~~about~~ 20 ppm oxygen.

16. The method according to claim 15, wherein steps (iv) – (vi) are repeated 1–10 times.

17. The method according to claims 15 or 16, wherein the base layer electroconductive paste comprises from ~~about~~ 1 to ~~about~~ 5 wt. % of adhesion promoter, preferably from ~~about~~ 2 to ~~about~~ 4 wt. %, more preferably ~~about~~ 3 wt. % of adhesion promoter.

18. The method according to any of claims 15-17, wherein the top layer electroconductive paste comprises from ~~about~~ 0.25 to ~~about~~ 1.25 wt. % of adhesion promoter, preferably from ~~about~~ 0.75 to ~~about~~ 1.25 wt. %, more preferably ~~about~~ 1 wt. % of adhesion promoter.

19. The method according to any of claims 15-18, wherein the adhesion promoter is bismuth oxide.

20. The method according to any of claims 15-19, wherein the nitrogen atmosphere comprises from ~~about~~ 1 to ~~about~~ 10 ppm oxygen, preferably from ~~about~~ 1 to ~~about~~ 3 ppm oxygen.

21. The method according to any of claims 15-20, wherein the step of subjecting the deposited base layer electroconductive paste to a temperature, and the step of subjecting the deposited layers to a temperature, includes heating from room temperature to ~~about~~ 950°C for ~~about~~ 5 to ~~about~~ 10 minutes and then cooling to room temperature.